FINAL REPORT

Estimated Exposure and Lifetime Cancer Incidence Risk from Plutonium Released from the 1969 Fire at the Rocky Flats Plant

Part of Task 3: Independent Analysis of Exposure, Dose,

and Health Risk to Offsite Individuals

August 1999

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EXECUTIVE SUMMARY

The Rocky Flats Environmental Technology Site is owned by the U.S. Department of Energy (DOE) and is currently contractor-operated by Kaiser-Hill Company. For most of its history, the site was called the Rocky Flats Plant (RFP) and was operated by Dow Chemical Company as a nuclear weapons research, development, and production complex. The RFP is located about 8 to 10 km (5–6 mi) from the cities of Arvada, Westminster, and Broomfield, Colorado, and 26 (16 mi) km northwest of downtown Denver, Colorado.

Through a 1989 Agreement in Principle between DOE and the State of Colorado, DOE provided the State with funding and technical support for health-related studies. The purpose of the Historical Public Exposures Studies on Rocky Flats is to estimate exposure to nearby residents from past toxic and radioactive releases from the plant.

This report documents fate and transport calculations and lifetime cancer incidence risk estimates for inhalation of plutonium¹ released during the glove box fire that occurred on May 11, 1969. Risk estimates are reported in terms of probability distributions that reflect the uncertainty in the calculation. This report presents estimates of time-averaged plutonium airborne concentrations at selected receptor locations within the model domain. Lifetime cancer incidence risks are then calculated for hypothetical individuals residing in the model domain who inhale plutonium. It provides a detailed discussion of the exposure scenarios that characterize behavior and physical attributes of hypothetical individuals. This report describes atmospheric transport modeling and uncertainty estimates, summarizes atmospheric transport calculations and risk estimates made in Phase I, and provides an overview of the source term developed for Phase II and documented in Voillequé (1999a). It also summarizes lifetime cancer incidence risks using risk coefficients and associated uncertainty developed by Grogan et al. (1999).

Summary of the Building 776/777 Glove Box Fire. On May 11, 1969, at about 2 p.m., a plutonium metal briquette stored in an open can in a glovebox spontaneously ignited. At 2:27 p.m., an alarm was received at the fire station. Two minutes later, when the captain and three firemen arrived at the west end of the building, there were flames 18 in. above the glovebox line. At 2:29 p.m., the firemen reported two loud noises and observed fireballs, presumably because of rapidly burning gases. Using experience gained fighting the 1957 fire, the captain directed that water be used to fight the fire. The fire spread along the north foundry glovebox line, but it was prevented from moving into the north machining glove boxes by a barrier. It then spread along the north-south conveyer glovebox line; when fire was observed in that area (2:50 p.m.), a loud noise was heard and firemen felt vibrations on the second floor of the building. Between 3:20 and 4:10 p.m., smoke was observed coming from the roof and exhaust vents. The roof was sprayed with water and watched until after 5 p.m. The fire was not considered contained until 6:40 p.m. The fire was considered to be "extinguished" by 8 p.m., and a fire watch was established at that time. Several small fires recurred during the night and the following morning.

Review of Phase I Evaluation of the 1969 Fire. ChemRisk used the limited air sampling and meteorological data collected during the fire to reconstruct the releases during the 1969 fire. The INtegrated PUFF dispersion code (INPUFF2), version 2 (<u>Petersen and Lavdas</u> 1986) was used to

¹ In this context, the word plutonium means weapons grade plutonium, which consists primarily of ²³⁹Pu (≈ 93.8%), ²⁴⁰Pu (≈5.8%), and ²⁴¹Pu (≈0.36%) by weight percent. Specific activity of weapons grade plutonium is 0.072 Ci g⁻¹.

model the atmospheric dispersion and deposition of the released plutonium. The code was used to estimate release rates that were consistent with the measured air concentrations.

Three sources of uncertainty were accounted for in the release estimates: uncertainty in the use of the INPUFF2 model; uncertainty associated with the air monitoring sampling devices and analytic techniques; and uncertainty associated with the limited time resolution and number of data points. The total estimated release of plutonium from the fire was 2.8 mCi. The upper and lower bounds of the 95% confidence interval about the median estimate were 56 mCi and 0.14 mCi, respectively.

ChemRisk predicted two contaminant plumes that extended east (Plume C) and southwest (Plume D) from the plant. Preliminary cancer risk estimates were also presented in Phase I of the study. A risk coefficient of 7.3% Sv⁻¹ was used based on recommendations from the International Commission on Radiological Protection (ICRP) (ICRP 1990). This risk coefficient includes fatal and nonfatal cancers and severe hereditary effects. The risk associated with the maximum exposed individual had a geometric mean of 1×10^{-7} with 2.5% and 97.5% values of 6×10^{-9} to 3×10^{-6} , respectively.

Phase II Release Estimates for the 1969 Fire. Relatively little plutonium was released to the atmosphere during the fire because of (a) the captain's decision to use water to fight the fire, (b) the persistence of the firemen, who made repeated entries into the building, and (c) the exhaust systems containing multiple sets of nonflammable high-efficiency particulate air (HEPA) filters were damaged but remained intact.

Some of the plutonium releases were measured. The main exhaust system samplers, operated between May 9 and 15, indicated that ~200 μ Ci (2.8 mg) was released via that exhaust. Measurements of surface contamination on the roof indicated that most of the release was via the Booster System #1 exhaust duct. The release from Booster System #1 was estimated to be in the range of 140 to 900 mg of plutonium. This release is much larger than the release from the room air exhaust. Some smoke was released through the open doors when the firemen entered the building at about 2:30 p.m., but that amount is also likely to have been much smaller than the release from Booster System #1. Total releases were estimated to range from 0.14 to 0.9 g of plutonium (0 and 100th percentiles). The particle size of the released plutonium was not measured. However, because most of the release had penetrated through four sets of HEPA filters, it is reasonable to believe that the particles were relatively small. Releases were postulated to occur over a 6-hour period. Model simulations were performed for 15 hours to allow the plume to completely dissipate from the model domain.

Environmental Transport Modeling. Five atmospheric transport models, ranging from a simple straight-line Gaussian plume model to a complex terrain model, were evaluated for use in this study (Rood 1999a). Models were compared to tracer measurements taken in the winter of 1991 at Rocky Flats. The results of this evaluation indicated no one model clearly outperformed the others. However, the puff trajectory models (Regional Atmospheric Transport Code for Hanford Emission Tracking [RATCHET], TRIAD, and INPUFF2) generally had lower variability and higher correlation to observed values compared to the other models. The RATCHET model was chosen for these calculations because it incorporates spatially varying meteorological and environmental parameters. Additionally, the model includes modules that perform random sampling of the meteorological parameters, allowing for Monte Carlo analysis of uncertainty.

The model domain encompassed a 2200-km^2 area (50 km north-south \times 44 km east-west). The domain extended 28 km south, 12 km west, 22 km north, and 32 km east from the RFP. Most of the Denver metropolitan area and the city of Boulder were included in the domain.

Meteorological data from RFP for the time of the fire were limited to the information in the letter to Dr. Roy Cleare, Executive Director of the Colorado Department of Health, dated March 20, 1970. These data were supplemented with data from Denver Stapleton International Airport and Jefferson County Airport. Hourly stability classes were calculated separately for the RFP, Denver Stapleton International Airport, and Jefferson Count Airport meteorological recording stations using the general classification scheme discussed in Pasquill (1961), Gifford (1961), and Turner (1964). This typing scheme employs seven stability categories ranging from A (extremely unstable) to G (extremely stable) and requires estimates of cloud cover and ceiling height. Cloud cover and ceiling height data for both stations were assumed to be the same and were obtained from the Denver Stapleton International Airport data.

The RATCHET simulation considered no plume rise from the release. This assumption was based on eyewitness accounts of the smoke plume and the fact that the effluent was discharged through inverted "J" vents that directed flow down toward the roof.

Treatment of Uncertainty. Risk estimates were reported as probability distributions that reflect our current state of knowledge about transport modeling, source term, dose and risk. They do not represent the probability of a seeing a health effect within the population of potential receptors. Uncertainty estimates for atmospheric transport modeling employed the Monte Carlo sampling features of the RATCHET code, which considered uncertainty in the wind speed, wind direction, Monin-Obukhov scaling length, and mixing height. This allowed for mass balance of material within the model domain for each of the 1000 Monte Carlo trials performed. Monte Carlo techniques were used to propagate atmospheric model prediction uncertainty through to the final risk calculations.

Predicted Air Concentrations. Distributions of 15-hour average concentrations for 15 receptor locations were described in terms of the 5th, 50th, and 95th percentile values of the distribution of predicted concentration values. These statistics were used to describe the concentration distributions and were not used in the risk calculation. The actual distributions comprising 1000 RATCHET realizations were used instead to calculate plutonium intake and risk to the receptors. The 15 receptor locations chosen for risk calculations represented individuals from each of the major population centers in addition to receptors placed at locations of high concentration in the model domain. Fifteen-hour average concentration values in the model domain ranged from minimum of 0.012 to a maximum of 31 fCi m⁻³ at the 5% level, and from 0.003 to 0.67 pCi m⁻³ at the 95% level.

Exposure Scenarios. The risk that a person experienced depends upon a number of factors, such as

- Where the person lived and worked in relation to the RFP
- Did the person live near the RFP during the 1969 fire
- Lifestyle (that is, did the person spend a great deal of time outdoors or doing heavy work on a farm)
- Age and gender of the person.

To consider these features of a person's life, we developed profiles (or exposure scenarios) of hypothetical, but realistic, residents of the RFP area for which representative risk estimates could be made. Risks were calculated for seven hypothetical exposure scenarios. These scenarios incorporate typical lifestyles, ages, genders, and lengths of time in the area. They can help individuals determine risk ranges for themselves by finding a lifestyle profile that most closely matches their background. The scenarios were not designed to include all conceivable lifestyles of residents who lived in this region during the time of RFP operations. Rather, they provide a range of potential profiles of people in the area.

The seven exposure scenarios were distributed at 15 locations within the model domain. Receptors were placed in major population centers and at points where the maximum concentrations in the model domain were observed. Scenarios included a rancher, housewife, infant, child, student, and laborer.

We only considered the inhalation pathway in this evaluation. We made this decision based on Phase I results that showed soil ingestion and inhalation of resuspended plutonium were minor pathways when considering the long-term exposure to Rocky Flats effluent (<u>ChemRisk</u> 1994a).

Plutonium Risk Coefficients. Lifetime cancer incidence risk coefficients (risk per unit intake) with uncertainty for plutonium were developed by <u>Grogan et al.</u> (1999) for the four critical organs: lung, liver, bone surface, and bone marrow (leukemia). Where feasible, gender- and agespecific risk coefficients were determined. Risk coefficients were reported for three different particle size distributions having geometric mean values of 1 μm, 5 μm, and 10 μm activity median aerodynamic diameter and a geometric standard deviation of 2.5 in all cases. For this assessment, all particles were assumed to be in the 1-μm range.

Incremental Lifetime Cancer Incidence Risk. Incremental lifetime cancer incidence risks were expressed in terms of percentiles of the cumulative density function. The receptor with the maximum total (all organs) risk in the model domain was the laborer located at the west entrance to the RFP. The risk to this receptor was 1.2×10^{-8} at the 25% level, 8.6×10^{-8} at the 50% level, and 4.9×10^{-7} at the 97.5% level. Using this receptor scenario as an example, the uncertainty in these risk estimates may be interpreted as follows:

- There is a 95% probability that the incremental lifetime cancer incidence risk was between 1.2×10^{-8} (2.5% value) and 4.9×10^{-7} (97.5% value).
- There is a 2.5% probability that the incremental lifetime cancer incidence risk was greater than 4.9×10^{-7} (97.5% value) and a 2.5% probability the risk was lower than 1.2 $\times 10^{-8}$ (2.5% value).

We can also interpret this to mean that given an exposure history and lifestyle similar to the laborer, there is a 97.5% probability that the model-predicted number of cancer cases attributed to inhalation of plutonium originating from the 1969 fire release would be no greater than 5 persons in a population of 10 million similarly exposed individuals. The organ with the greatest risk was the lung, followed by the liver, bone, and bone marrow.

An almost infinite number of possible exposure scenarios can be defined; in most cases, the risks associated with each scenario will differ. However, the maximum risks will probably be bounded by the risks associated with the laborer scenario. This scenario may be considered the maximum exposed individual in the model domain because the laborers were placed at the point

of highest concentration outside the RFP buffer zone. The calculated risks were within the U.S. Environmental Protection Agency point of departure for acceptable lifetime cancer incidence risk of 1 in 1,000,000 to 1 in 10,000 people.

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ACRONYMS

AED aerodynamic equivalent diameter
AMAD activity median aerodynamic diameter
ASCOT Atmospheric Studies in Complex Terrain

CDPHE Colorado Department of Public Health and Environment

DOE U.S. Department of Energy

EPA U.S. Environmental Protection Agency

GM geometric mean

GSD geometric standard deviation

HAP Health Advisory Panel

HEPA high-efficiency particulate air (filter)

ISC Industrial Source Complex Short Term Version 2 INPUFF2 INtegrated PUFF dispersion (code) version 2

LET linear energy transfer

RAC Radiological Assessments Corporation¹

RATCHET Regional Atmospheric Transport Code for Hanford Emission Tracking

RBE relative biological effectiveness

RFP Rocky Flats Plant

TIC time-integrated concentration TLLa total long-lived alpha activity

TRAC Terrain Responsive Atmospheric Code

USGS U.S. Geological Survey UTM universal transverse mercator

WVTS Winter Validation Tracer Study

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¹ In 1998 *Radiological Assessments Corporation* changed its name to *Risk Assessment Corporation*. For consistency throughout the project, all reports were published by *Radiological Assessments Corporation*.

INTRODUCTION

The Rocky Flats Environmental Technology Site is owned by the U.S. Department of Energy (DOE) and is currently contractor-operated by Kaiser-Hill Company. For most of its history, the site was called the Rocky Flats Plant (RFP) and was operated by Dow Chemical Company as a nuclear weapons research, development, and production complex (Figure 1). The RFP is located on approximately 2650 ha (6500 acres) of Federal property, about 8–10 km from the cities of Arvada, Westminster, and Broomfield, Colorado, and 26 km northwest of downtown Denver, Colorado. The original 156-ha (385-acre) main production area is surrounded by a 2490-ha (6150-acre) buffer zone that now delineates the RFP boundary.

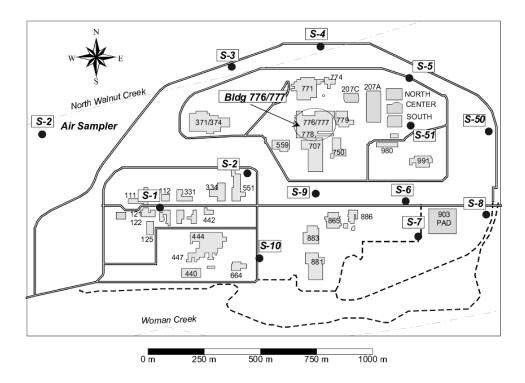


Figure 1. Main production area of the Rocky Flats Plant as it appeared in 1990. Originally, the buildings were identified with two-digit numbers. Later, a third digit was added. The production area, now sometimes called the industrial area, is surrounded by a security perimeter fence. Building 776/777 identified in the northern part of the facility was the primary release point from the fire. Placement of air samplers is based on air sampler locations and numbers that existed before 1973. See Chapter III and Appendix B in Rope et al. (1999) for maps of air sampler locations as they existed with respect to past features.

Through a 1989 Agreement in Principle between the DOE and the State of Colorado, DOE provided the State with funding and technical support for health-related studies. The purpose of the Historical Public Exposures Studies on Rocky Flats is to estimate exposure to nearby residents from past toxic and radioactive releases from the plant. The Colorado Department of Public Health and Environment (CDPHE) first invited a national panel of experts to help design

the health studies. Because of intense public concern about Rocky Flats contamination among Denver metropolitan area residents following a Federal Bureau of Investigation raid of Rocky Flats in June 1989, the panel decided to stress public involvement and to separate the research into two major phases conducted by two different contractors to enhance accountability and credibility.

Phase I of the study was performed by ChemRisk (a division of McLaren/Hart, Environmental Engineering). In Phase I, ChemRisk conducted an extensive investigation of past operations and releases from the RFP. The Phase I effort identified the primary materials of concern, release points and events, quantities released, transport pathways, and preliminary estimates of dose and risk to offsite individuals. The conclusions from Phase I were released in a public summary document by the Health Advisory Panel (HAP) (HAP 1993); a series of task reports by ChemRisk (ChemRisk 1994a, 1994b, 1994c, 1994d); and several articles in the journal Health Physics.

Radiological Assessments Corporation (RAC) was awarded the contract to conduct Phase II of the study, which is an in-depth investigation of the potential doses and risks to the public from historical releases from Rocky Flats. Recommendations for work to be performed in Phase II are outlined in the Phase I summary document <u>HAP</u> (1993).

This report documents fate and transport calculations for plutonium¹ released during the fire that occurred in Building 776/777 on May 11, 1969. The fire, which started in a glovebox, resulted in plutonium releases from the ventilation system within the building. However, the filtration system was not breached. We estimated time-averaged plutonium airborne concentrations at different receptor locations within the model domain and calculated lifetime cancer incidence risks for hypothetical individuals residing in the model domain who inhaled airborne plutonium. This report provides a detailed discussion of the exposure scenarios that characterize behavior and physical attributes of hypothetical individuals. It also describes details of atmospheric transport modeling and uncertainty estimates, summarizes atmospheric transport calculations and risk estimates made in Phase I, and provides an overview of the source term developed for Phase II and documented in Voillequé (1999a). This report also summarizes lifetime cancer incidence risks using risk coefficients and associated uncertainty developed by Grogan et al. (1999) and reviews and discusses soil, vegetation, and air monitoring data useful for model validation.

SUMMARY OF THE GLOVE BOX FIRE IN BUILDING 776/777

A detailed accounting of the chronology and release estimates from the fire that occurred in Building 776/777 on May 11, 1969, is documented in <u>Voillequé</u> (1999a). This section provides a brief summary of the events of the fire. Release estimates are given in a later section.

The fire in Buildings 776/777 in May 1969 was a landmark event in the history of the RFP. Unlike 1957 and earlier years, when the word plutonium was never mentioned, the functions of the RFP had been revealed before the 1969 fire, and the potential impact of the fire was understood by many. Following the fire, the Atomic Energy Commission conducted a detailed investigation of the event. Independent scientists also investigated the Rocky Flats environment.

¹ In this context, the word plutonium means weapons grade plutonium, which consists primarily of 239 Pu (≈93.8%), 240 Pu (≈5.8%), and 241 Pu (≈0.36%) by weight percent. Specific activity of weapons grade plutonium is 0.072 Ci g⁻¹.

Poet and Martell (1972) published measurements of plutonium soil contamination that showed releases had occurred during previous years of operation. Their findings led to revelations about the improper storage of plutonium-contaminated oil in the 903 Area and about failure of the effluent sampling system during the 1957 fire in Building 771. Estimates of the releases from the 903 Area and during the 1957 fire are presented in other Rocky Flats dose reconstruction reports (Weber et al. 1998; Voillequé 1999b).

On May 11, 1969, at about 2 p.m., a plutonium metal briquette stored in an open can in a glovebox spontaneously ignited. At 2:27 p.m., an alarm was received at the fire station. Two minutes later, when the captain and three firemen arrived at the west end of the building, there were flames 18 in. above the glovebox line. At 2:29 p.m., the firemen reported two loud noises and observed fireballs, presumably because of rapidly burning gases. Using experience gained fighting the 1957 fire, the captain directed that water be used to fight the fire. The fire spread along the north foundry glovebox line, but it was prevented from moving into the north machining gloveboxes by a metal accountability barrier. It then spread along the north-south conveyer glovebox line; when fire was observed in that area (2:50 p.m.), a loud noise was heard and firemen felt vibrations on the second floor of the building. Between 3:20 and 4:10 p.m., smoke was observed coming from the roof and exhaust vents. The roof was sprayed with water and watched until after 5 p.m. The fire was not considered contained until 6:40 p.m. The fire was considered to be "extinguished" by 8 p.m., and a fire watch was established at that time. Several small fires recurred during the night and the following morning.

Plutonium metal briquettes were produced from scrap metal in an area adjacent to the production glovebox line and were stored in the production area. The plutonium storage glovebox and other glove boxes contained tons of flammable Benelex and Plexiglas. Benelex, composed of wood fiber and plastic, was used to shield against neutron radiation. In addition to the flammable shielding and glovebox window material, it was reported that there were also combustible oily rags in the area where the fire started. The glovebox and building ventilation systems provided a continuing supply of oxygen for combustion. Heat sensors did not function as designed because of the large amount of shielding in the storage cabinets and because the plutonium storage cans were not compatible with the sensor design. Building 776/777 was largely open, with few fire breaks and no installed sprinkler system. The water used to fight the fire was brought in using hoses.

REVIEW OF THE PHASE I EVALUATION OF THE 1969 FIRE

To analyze the 1969 fire, ChemRisk divided it into the following two release periods (ChemRisk 1994c):

- From the start of the fire at 2:30 p.m. until 8:00 p.m. on May 11, 1969, when the fire was extinguished (with the exception of small fires in the North Foundry line that continued to reoccur throughout the night)
- From 8:00 p.m. on May 11, 1969, until 10:00 a.m. on May 12, 1969, when the fire was assumed to be completely extinguished.

In the absence of direct measurements and because the main filter plenum and booster systems were largely intact, ChemRisk assumed that the majority of particles emitted during the fire were very small in size (less than 1 micron). ChemRisk used the limited air sampling and

meteorological data collected during the fire to reconstruct the releases during the 1969 fire. The INtegrated PUFF dispersion (INPUFF2) (Petersen and Lavdas 1986) code was used to model the atmospheric dispersion and deposition of the released plutonium. The code was used to estimate release rates that were consistent with the measured air concentrations. The majority of wind observations on which the INPUFF2 modeling were based were only taken once an hour; therefore, the data were "smoothed" by assigning an intermediate wind direction for a portion of each hour when the wind shifted between two consecutive hours.

For the analysis, it was assumed that removal of the fine particles from the air by deposition to the ground played a minor role. A zero settling velocity and a deposition velocity of 0.1 cm s⁻¹ were assumed, which is consistent with submicron-size particles. However, because the actual particle size distribution was not known, an uncertainty factor of 3 was incorporated into the dose calculations. Precipitation scavenging was not taken into account for the 1969 fire because no precipitation was recorded in the region around that time. The stack height was accounted for in the modeling.

Three sources of uncertainty were accounted for in the release estimates: uncertainty in the use of the INPUFF2 model; uncertainty associated with the air monitoring sampling devices and analytic techniques; and uncertainty associated with the limited time resolution and number of data points.

An average plutonium release rate of $0.05~\mu Ci~s^{-1}$ and $0.02~\mu Ci~s^{-1}$ was determined for the first and second release periods, respectively. The total estimated release of plutonium from the fire was 2.8~mCi. The upper and lower bounds of the 95% confidence interval about the median estimate were 56~mCi and 0.14~mCi, respectively.

ChemRisk predicted two contaminant plumes extended east (Plume C) and southwest (Plume D) from the plant. The inhalation dose estimates for plutonium released during the 1969 fire are presented in Table 1 as a function of distance along the centerline of the two plumes. Pathway-specific and annual total doses resulting from the plutonium deposited during the 1969 fire were also calculated. The total annual doses were 4 orders of magnitude lower than the inhalation doses reported in Table 1.

Table 1. Phase I Dose and Risk Estimates for Airborne Releases of Plutonium
during the 1969 Fire

Location – distance from plant	Inhalation dose ^a (Sv)	Risk estimate ^b
Plume C – 3 mi	$4.5 \times 10^{-6} (4.6)$	$3 \times 10^{-7} (2 \times 10^{-8} \text{ to } 7 \times 10^{-6})$
Plume C – 5 mi	$1.7 \times 10^{-6} (4.6)$	$1 \times 10^{-7} $ (6 × 10^{-9} to 3 × 10^{-6})
Plume C – 8 mi	$7.6 \times 10^{-7} (4.6)$	$6 \times 10^{-8} (3 \times 10^{-9} \text{ to } 1 \times 10^{-6})$
Plume $D - 3 \text{ mi}$	$2.1 \times 10^{-7} (4.6)$	$2 \times 10^{-8} \ (7 \times 10^{-10} \ \text{to} \ 3 \times 10^{-7})$
Plume D – 5 mi	$7.6 \times 10^{-8} (4.6)$	$6 \times 10^{-9} (3 \times 10^{-10} \text{ to } 1 \times 10^{-7})$
Plume D – 8 mi	NA ^c	NA

^a From Appendix L, ChemRisk (1994d); geometric mean (geometric standard deviation)

Preliminary cancer risk estimates were also presented in Phase I of the study. A risk coefficient of $7.3\%~Sv^{-1}$ was used based on recommendations from the International Commission on Radiological Protection (ICRP) (ICRP 1990). This risk coefficient includes fatal and nonfatal

^b From Figure 5-3, ChemRisk (1994d); geometric mean (2.5 percentile to 97.5 percentile)

 $^{^{}c}$ NA = not applicable.

cancers and severe hereditary effects. The risk estimates associated with inhalation exposure during the 1969 fire are given in <u>Table 1</u>.

PHASE II RELEASE ESTIMATES FOR THE 1969 FIRE

Relatively little plutonium was released to the atmosphere during the fire. This result is attributed to several factors. First, the captain's decision to use water to fight the fire was a crucial one. In addition, the persistence of the firemen, who made repeated entries into the building, was very important to controlling the fire and limiting the release. A third factor was that the most important exhaust systems contained multiple sets of nonflammable high-efficiency particulate air (HEPA) filters. The room ventilation air was exhausted through a single set of HEPA filters. However, the booster exhaust and dry air systems each had four to six sets of HEPA filters in sequence. Filters in Booster System #2 were plugged by smoke during the early stages of the fire, and the main exhaust flow was carried along the Booster System #1 exhaust path via the conveyer glovebox line. This was the main release path. After HEPA filtration, the booster system exhausts were discharged through inverted-J ducts near the surface of the building's roof. The roof was contaminated as a result. Plutonium was also tracked out of the building by the firemen who came out to obtain fresh air supplies.

Some of the plutonium releases were measured. The main exhaust system samplers, operated between May 9 and 15, indicated that ~200 μ Ci (2.8 mg) of plutonium was released via that exhaust. Booster system sampling was incomplete because of a power failure. Measurements of surface contamination on the roof indicated that most of the release was via the Booster System #1 exhaust duct. Later investigation showed that there was damage to all four stages of HEPA filtration in the Booster System #1 exhaust line. Damage to the fourth stage was limited to the upper section. The release from Booster System #1 was estimated to be in the range of 140 to 900 mg of plutonium. This release is much larger than the release from the room air exhaust. Some smoke was released through the open doors when the firemen entered the building at about 2:30 p.m., but that amount is also likely to have been much smaller than the release from Booster System #1.

The particle size of the released plutonium was not measured. However, because most of the release had penetrated through four sets of HEPA filters, it is reasonable to believe that the particles were relatively small.

Release quantities are detailed in <u>Voillequé</u> (1999a) and are summarized in <u>Table 2</u>. Releases are reported in terms of percentiles of the cumulative probability density function for each 15-minute period during the release event.

ENVIRONMENTAL MONITORING OF PLUTONIUM

Historical environmental monitoring data relevant to assess contaminant emissions from the RFP are evaluated in Rope et al. (1999). This section briefly reviews data that are pertinent to the release of plutonium from the 1969 fire event, emphasizing potential model validation data. This limits our discussion to measurements in ambient air and vegetation. Most soil measurement data were taken after 1970; therefore, evidence of deposition from the 1969 fire is obscured by deposition from 903 Area releases and the cumulative deposition from routine releases. In

general, the *offsite* air monitoring and vegetation data were also found to be of little use for validating model predictions because

- Offsite air monitoring data are available for total long-lived alpha activity (TLLa) only. Most of the data were below the minimum detectable activity and have large counting errors, which make it difficult to determine reliable estimates for background or fallout plutonium concentrations. Furthermore, interference from 903 Area releases make reliable estimates from the 1969 fire difficult.
- Only very limited vegetation data are available for model validation, and the dominance of releases from the 903 Area obscures the contribution from the 1969 fire.

Table 2. Summary of May 11, 1969, Fire Plutonium Source Term Reported in Voillequé (1999a)

	•	(2>>> 0)	
_	Release quantities (mCi)		
Time	5%	50%	95%
2:00–2:15 p.m.	0.163	0.466	0.773
2:15-2:30 p.m.	0.163	0.466	0.773
2:30-2:45 p.m.	0.163	0.466	0.773
2:45-3:00 p.m.	0.163	0.466	0.773
3:00-3:15 p.m.	0.651	1.865	3.090
3:15–3:30 p.m.	0.651	1.865	3.090
3:30–3:45 p.m.	0.651	1.865	3.090
3:45–4:00 p.m.	0.651	1.865	3.090
4:00–4:15 p.m.	1.139	3.264	5.408
4:15–4:30 p.m.	1.139	3.264	5.408
4:30–4:45 p.m.	1.139	3.264	5.408
4:45–5:00 p.m.	1.139	3.264	5.408
5:00–5:15 p.m.	0.814	2.331	3.863
5:15–5:30 p.m.	0.814	2.331	3.863
5:30–5:45 p.m.	0.814	2.331	3.863
5:45–6:00 p.m.	0.814	2.331	3.863
6:00–6:15 p.m.	0.325	0.932	1.545
6:15–6:30 p.m.	0.325	0.932	1.545
6:30–6:45 p.m.	0.325	0.932	1.545
6:45–7:00 p.m.	0.325	0.932	1.545
7:00–7:15 p.m.	0.163	0.466	0.773
7:15–7:30 p.m.	0.163	0.466	0.773
7:30–7:45 p.m.	0.163	0.466	0.773
7:45–8:00 p.m.	0.163	0.466	0.773
Total	13	37	62

Ambient Air Monitoring

A detailed review and analysis of plutonium monitoring in air around Rocky Flats is documented in Rope et al. (1999). At the time of the 1969 fire, air monitoring was performed by the site contractor and several other independent agencies. Up until 1970, air samplers were analyzed for TLLa only. The RFP contractor had a total of 12 onsite ambient air monitoring stations in 1969 (Figure 1). These 12 samplers (designated S-1 through S-10, S-50, and S-51) were operating at the time of the 1969 fire. Filters were collected from the samplers on weekday mornings and were counted for gross alpha radioactivity. Each week was represented by four 24hour samples (filters collected Tuesday, Wednesday, Thursday, and Friday) and one 72-hour weekend sample (filter collected Monday). Because the fire occurred on a Sunday (May 11, 1969), the 72-hour weekend sample would have reflected the releases during the fire. Copies of the handwritten daily results sheets titled, "Health Physics On Site, Site Survey Routine Air Sample Results," were retrieved from the Denver Federal Records Center by RAC and transcribed into a spreadsheet for analysis. For samples collected over periods longer than 1 day, the total net counts per minute was divided by the number of days of sampling so that all results were estimates of daily counts. The daily counts were converted into air concentrations, reported in femtocuries per cubic meter (fCi m⁻³), taking into account analytic and measurement errors, flow rate, and sampling time. These data are potentially useful for validating releases from the 1969 fire. However, model comparisons are contingent upon isolating concentrations attributed to the fire, from the 3-day weekend sample counts, and accounting for contributions from other sources of plutonium that included the 903 Area.

In addition to the onsite air samplers, the site had nine offsite air samplers operating during 1969. These were located at Coal Creek (S-11), Marshall (S-13), Boulder (S-15), Lafayette (S-16), Broomfield (S-17), Wagner (S-18), Golden (S-20), Custom House in Denver (S-23), and Westminster (S-25). Again, measurements of TLLa only were made. Although these were continuous air samplers, the samplers were programmed to operate for only 5 minutes every hour. Thus, the actual operating time was typically around 14 hours per week, which was less than 10% of the total time period. The short sample time and low flow rate of these samplers produced a small daily sample volume (~40 to 50 m³), which was less than the onsite daily sample volumes (81.5 m³). The small sample volume combined with the variable counter background rate resulted in poor sensitivity of the analysis. It is considered unlikely that these data will be useful for any validation efforts.

Vegetation Monitoring

Routine offsite vegetation sampling data for 1969 were reported in the Environmental Survey Reports generated by Dow Chemical Company. Samples, collected at approximately 100 locations, were analyzed for TLLa (<u>Hammond</u> 1969). The Environmental Survey Reports present the average and maximum values for all samples collected within a given distance from the plant, for example, <3 mi, 3–18 mi, >18 mi.

Special vegetation sampling was conducted following the May 11, 1969, fire. Twenty-two samples were collected from 11 offsite locations (Figure 2). The samples were analyzed for TLLa and plutonium. These data are also reported as averages within given distances from Rocky Flats. As noted in Rope et al. (1999), it is unclear whether onsite values are included with

these data. The onsite vegetation data related to the 1969 fire were recorded in handwritten logbooks (<u>Dow</u> 1953–1971), and reported by <u>Hammond</u> (1969). Figure 3 is based on a hand-drawn map of the onsite sampling grid on which selected plutonium concentrations for approximately 20 locations were noted. The importance of the 903 Area as a major source of contamination is evident from these data, which made it difficult to isolate the impact of the 1969 fire.

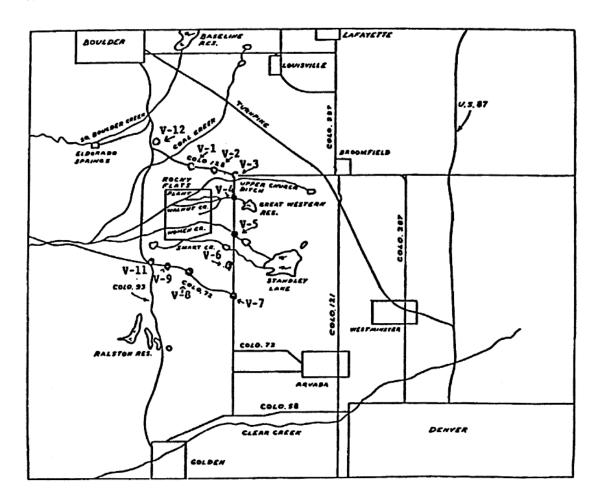


Figure 2. Map of offsite locations for special vegetation sampling following the 1969 fire measured in vegetation samples collected after the May 1969 fire in Building 776/777 (reproduced from <u>Hammond</u> 1969).

ENVIRONMENTAL TRANSPORT MODELING

Offsite exposure to plutonium from releases resulting from the May 1969 glove box fire in Building 776/777 were investigated in Phase I and are summarized in a previous section of this report. Airborne releases were considered to be the principal transport pathway and inhalation the major pathway of exposure.

Atmospheric releases of plutonium as a result of the glove box fire primarily occurred from the inverted "J" roof vents. This section describes our approach to estimating atmospheric

dispersion of plutonium released from this event and the uncertainty associated with concentration estimates in the model domain. Our approach to this calculation involved first estimating the plume trajectory based on the available meteorological data. Next, the stochastic source term developed by Voillequé (1999a) was coupled with the atmospheric dispersion model to generate concentration isopleths in the model domain, incorporating uncertainties in the dispersion process. Distributions of airborne concentrations were then used with exposure scenarios and plutonium inhalation risk coefficients to calculate incremental lifetime cancer incidence risk for hypothetical receptors in the model domain.

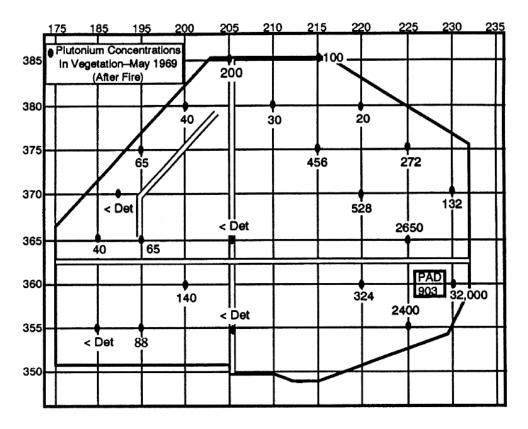


Figure 3. Map of onsite locations for special vegetation sampling following the 1969 fire measured in vegetation samples collected after the May 1969 fire in Building 776/777 (reproduced from <u>Hammond</u> 1969).

Atmospheric Model Selection

Five atmospheric transport models considered for use in this study were evaluated in <u>Rood</u> (1999a): (1) Terrain-Responsive Atmospheric Code (TRAC) (<u>Hodgin</u> 1991), (2) the Industrial Source Complex Short Term Version 2 (ISC) (<u>EPA</u> 1992), (3) Regional Atmospheric Transport Code for Hanford Emission Tracking (RATCHET) (<u>Ramsdell et al.</u> 1994), (4) TRIAD (<u>Hicks et al.</u> 1989), (5) and INPUFF2 (<u>Petersen and Lavdas</u> 1986). The model comparison study determined what models, if any, performed best in the Rocky Flats environs for a given set of modeling objectives.

Model evaluations were based on how well predictions compared with measured tracer concentrations taken during the Winter Validation Tracer Study (WVTS) (Brown 1991) conducted in February 1991 at the RFP. The WVTS consisted of 12 separate tests: 6 tests were conducted during nighttime hours, 4 during daytime hours, and 2 during day-night transition hours. For each test, an inert tracer (sulfur hexafluoride) was released at the RFP at a constant rate for 11 hours from a 10-m high stack located on the southern boundary of the RFP industrial area. Two sampling arcs, 8 and 16 km from the release point, measured tracer concentrations every hour for the last 9 hours of each test period. Seventy-two samplers were located on the 8-km arc, and 68 samplers were located on the 16-km arc. Predicted concentrations were then compared to the observed tracer concentrations at each of the samplers.

We acknowledge that the release conditions of the WVTS are substantially different from the glove box fire release conditions. Most notably, the release geometry (multiple release points on a building roof compared to a single point for the WVTS) and release temperature are different. Despite these shortcomings, the WVTS is the most complete site-specific data set available with which to evaluate atmospheric transport models. We can expect that an elevated, multiple-point release will increase the uncertainty in a model prediction, but we assumed that the relative performance among models was adequately characterized by the comparison with the WVTS data.

Modeling objectives for the comparison study were based on the premise that identifying hourly locations of individual receptors was unlikely. Instead, it was more likely to identify receptors (hypothetical or real) who were present at a fixed location for the duration of a release event. The minimum time scale of historical release events at RFP ranged from 6-10 hours to several days. Release events modeled for the WVTS were 9 hours in duration. If we assume the receptor is fixed for a time period of at least 9 hours, then the time-averaged concentration (9-hour average) is an appropriate modeling objective rather than comparing hourly average concentrations. Therefore, models were evaluated based on their performance in predicting timeaveraged concentrations at fixed sampler locations in the model domain (9-hour average concentration at each sampler paired with the corresponding predicted value). We also considered the arc-integrated concentration. The arc-integrated concentration was the 9-hour average ground-level concentration integrated across the 8- and 16-km sampling arc. The latter performance objective provides a measure of the vertical dispersion component of the models and the ground-level tracer mass, 8 and 16 km from the release point. Data sets for the timeaveraged concentration were limited to only those points where the predicted (C_n) and observed (C_0) concentration pair were greater than the time-averaged minimum detectable concentration.

Fifty percent of the time-averaged model predictions were within a factor of 4 of the observations. Predicted-to-observed ratios (C_p/C_o) ranged from 0.001 to 100 and tended to be higher at the 16-km arc than the 8-km arc. Geometric mean (GM) C_p/C_o ratios ranged from 0.64 (TRAC) to 1.5 (ISC), and geometric standard deviations (GSDs) ranged 4.4 (RATCHET) to 6.5 (ISC). The RATCHET model had the highest correlation coefficient for the 8-km (0.67) and 16-km (0.58) sampling arcs, followed by TRIAD and INPUFF2 (Figure 4). Qualitatively, the predictions made by the RATCHET model appear to best match the observations. The slope of the regression line was closest to that of the perfect correlation line (solid line in Figure 4).

Arc-integrated results showed INPUFF2 and TRIAD had the highest correlation coefficients, but correlation coefficients were not significantly different (at the 95% level) from the other

models. The ISC model tended to overpredict arc-integrated concentration, and the TRAC model showed the greatest variability.

The results reported in Rood (1999a) indicated no one model clearly outperformed the others. However, the RATCHET, TRIAD, and INPUFF2 models generally had lower variability (indicated by lower GSDs of C_p/C_o ratios) and higher correlation coefficients compared to those of ISC and TRAC models. It is desirable in a study such as this to choose a model that has the least amount of variability when comparing model predictions to observations. In addition, the model selected should have a level of complexity that is consistent with available data. The TRAC model is the most complex in terms of its treatment of the atmospheric dispersion process in complex terrain, but the study showed model performance was no better than the other models. In addition, we are lacking the meteorological data needed to fully use the capabilities of the TRAC model. The straight-line Gaussian plume model, ISC, tended to overpredict concentrations and was also limited to only one meteorological recording station in the model domain. Available meteorological data for this study period included two meteorological recording stations: one at the RFP and the other at Denver Stapleton International Airport. Therefore, it is desirable to use a model that may include multiple meteorological recording stations in the model domain. Using multiple meteorological recording stations allows for a spatially varying wind field in the model domain.

The RATCHET, INPUFF2, and TRIAD models performed comparably and were considered viable candidates for atmospheric dispersion estimates. Of these models, RATCHET and TRIAD were chosen for more detailed evaluation because both these models allow for spatially varying wind fields.

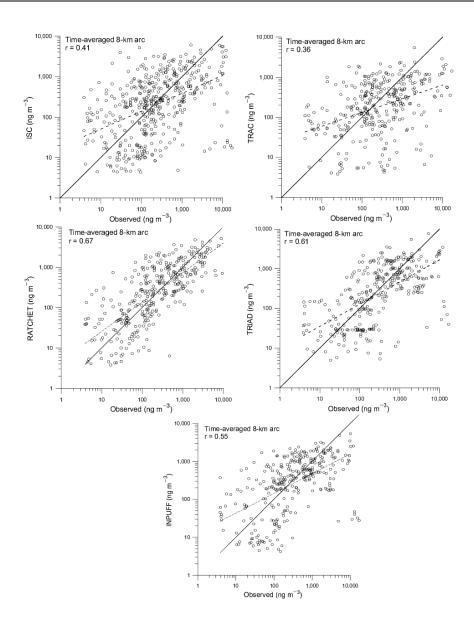


Figure 4. Nine-hour average observed concentrations as a function of predicted values for the five models compared using the WVTS data set. Correlation coefficients were for the log-transformed data. The solid line represents perfect correlation between predicted and observed values. The dashed line represents the log-transformed regression fit.

There are advantages and disadvantages to using either model in dispersion calculations. The TRIAD model is capable of incorporating meteorological data on a user-defined time scale, while RATCHET uses a fixed, 1-hour increment. Meteorological data from the RFP during the fire were reported every 15 minutes; therefore, the TRIAD model would appear better suited for the calculation because it could incorporate the resolution of the meteorological data. However, Denver Stapleton International Airport data were recorded every hour, so this advantage is lost in terms of predicting wind vectors at that distance. In addition, model comparisons using the WVTS data set showed that RATCHET performed as well, if not better than TRIAD, using

1-hour average meteorological conditions. RATCHET also has several other features that make it desirable, including

- Spatial varying surface roughness lengths and mixing heights
- Algorithms to compute plume depletion and deposition for fine particles are included (deposition must be computed outside the TRIAD codes)
- Random sampling routines that facilitate Monte Carlo calculations.

We chose the RATCHET model to perform the calculations based on its performance in the WVTS model comparison (Rood 1999a) and the features of the code stated previously. Features of the RATCHET model are summarized in Table 3.

Table 3. Features of the RATCHET Model

Feature	Representation in RATCHET
Domain area ^a	2200 km^2
Node spacing ^a	2000 m
Source term	Hourly release rates
Meteorological data	Hourly
Surface roughness	Spatially varying
Wind fields	$1/r^2$ interpolation (r = the radial distance from the observation)
Topographical effects	None explicit ^b
Wind profile	Diabatic
Stability	Spatially varying based on wind, cloud cover, and time of day
Precipitation	Spatially varying, three precipitation regimes with different precipitation rate distributions
Mixing layer	Spatially varying, based on calculated values for each meteorological station
Plume rise	Briggs' equation (Briggs <u>1969</u> , <u>1975</u> , <u>1984</u>)
Diffusion coefficients	Based on travel time and turbulence levels
Dry deposition	Calculated using resistance model
Wet deposition	Reversible scavenging of gases, irreversible washout of particles
Model time step	15 minute maximum, 15 second minimum
Output frequency	Hourly
Uncertainty	Options available for Monte Carlo simulation within the code

^a Modified from the original RATCHET specification for use at Rocky Flats.

Model Domain and Receptor Grid

The model domain (Figure 5) encompasses a 2200 km^2 area (50 km north-south \times 44 km east-west). The domain extends 28 km south, 12 km west, 22 km north, and 32 km east from the RFP. Most of the Denver metropolitan area and the city of Boulder are included in the domain.

Terrain differences are not a model input. However, topographical influence on the wind field may be accounted for by incorporating multiple meteorological stations in the model domain.

RATCHET was modified to accommodate hourly output. The original code output time-integrated concentrations daily.

The domain was limited in its western extent because few receptors are present there and most of the contaminant plumes traveled east and southeast of the plant.

RATCHET uses two modeling grids. Hourly meteorological records are used to estimate wind speed and direction, stability, and precipitation on the environmental grid in addition to surface roughness features. The concentration grid has spacing one-half that of the environmental grid. Ground-level concentrations and deposition are output at each of these grid nodes. The environmental grid was set at 23 nodes east-west and 26 nodes north-south, with a grid spacing of 2000 m. The concentration grid has 45 nodes east-west and 51 nodes north-south, with a spacing of 1000 m. The southwest corner of the model domain has the universal transverse mercator (UTM) coordinates 470850 E and 4387050 N. Release points are defined by distances (in kilometers) from a reference node. The reference node for the environmental grid was (7,15), and the reference node for the concentration gird was (13,29); they both have the UTM coordinates of 482850 E and 4415050 N.

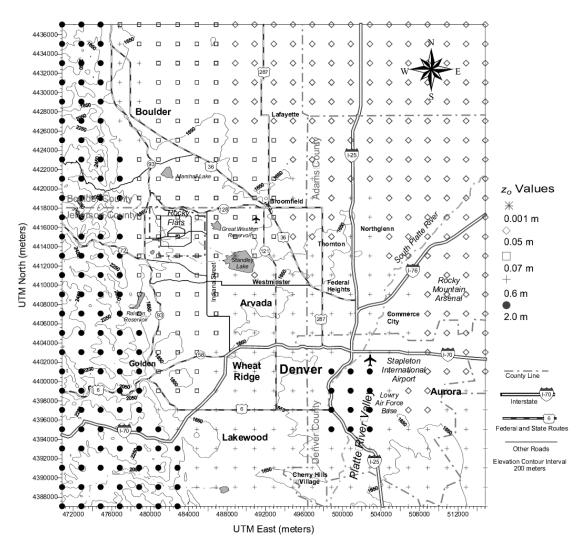


Figure 5. RATCHET environmental modeling grid and roughness length values (z_o) . Symbols represent grid nodes and the z_o value assigned to the node.

<u>Figure 5</u> was generated using U.S. Geological Survey (USGS) 7.5-minute digital elevation models. Topographic contours were based on an elevation grid spacing of 100 m. Major roadways and water features were digitized from USGS 1:100,000 digital line graphs.

Meteorology

Rocky Flats meteorological data for its operational period (1953–1989) are sporadic, incomplete, and of questionable integrity. Requests for meteorological data from the RFP were initially made by ChemRisk during Phase I of the project. ChemRisk was able to locate two letters from Dow Chemical to Dr. Roy Cleare, Executive Director of the Colorado Department of Health, dated March 20, 1970. These letters contained wind speed and direction for varying time increments during the 1957 and 1969 fire incidents. Computer diskettes containing wind speed, wind direction, and precipitation measurements from October 1968 to May 1969 were also obtained. These data were hourly observations taken approximately 15 minutes before the top of the hour and do not represent hourly average readings. Although these data appeared to be climatologically reasonable, no records of instrument calibration or audits of the information were found. Parameter resolution was very coarse (for example, wind direction resolution was 45 degrees). Original records, including the strip recording charts, were not located for the period from 1952–1983.

An extensive data search was initiated in 1994 by *RAC* researchers to locate missing data and interview personnel who were involved with measurements at the site. No new data were recovered, but several personnel reported problems with the recording instrumentation at the RFP (for example, the measured wind direction was off by 180 degrees). In 1983, a 61-m tower was constructed near the southern boundary of the RFP industrial area. Meteorological instrumentation was installed at 10-, 25-, and 61-m heights. These instruments were coupled with digital data recorders that allowed data to be taken continuously and processed and stored on a 15-minute interval. Operation of the tower began in 1984, and data recording adhered to strict quality assurance standards. Data from 1989–1993 were used in conjunction with data from Denver Stapleton International Airport to estimate annual average dispersion factors (Rood 1999b).

In 1994, the RFP hired a subcontractor to compile, screen, validate, and analyze historical climatological data (<u>DOE</u> 1995). A draft report issued in February 1995 contained monthly and annual summaries of wind speeds, wind directions, precipitation, temperature, and other parameters for the years 1953–1993. While these data are of interest and may be important for some aspects of modeling, they lacked the resolution required for detailed atmospheric transport modeling.

Except for the March 20, 1970, letter from Dow Chemical to Dr. Roy Cleare and the other electronic records, meteorological records from Rocky Flats were lacking. Other options were considered, such as using typical meteorological conditions for the month of May for years where reliable meteorological data were recorded (1984–present). However, while this technique is suitable for long-term dispersion estimates (as was done for the routine release assessment [Rood 1999b]), it was not viable for estimating conditions during a relatively brief event like the 1969 glove box fire because daily conditions were highly variable. Therefore, we had little recourse but to use the meteorological data provided in the Cleare letter despite its questionable nature. Data from Denver Stapleton International Airport and Jefferson County Airport covering

the period of interest (May 11–12, 1969) were also obtained and used in conjunction with the RFP data for air dispersion calculations. These data were instantaneous measurements, not hourly averages as was typical of all airport data before the Automatic Surface Observation Site system was installed at most major airports. The Denver Stapleton International Airport meteorological station is located 24 km east and 14 km south and the Jefferson Count Airport is located 8.4 km east and 2.5 km south from the center of the model domain (RFP). Data from both these stations included measurements of wind speed, wind direction, cloud cover, and precipitation. Jefferson County Airport was closed from 11:00 p.m. to 5:00 a.m. the following day. However, during the period of highest releases (2:00 p.m. to 8:00 p.m. May 11, 1969), the station was operating.

Data Processing

Meteorological data from the Cleare letter were obtained from the Phase I Task 6 report (ChemRisk 1994c, Appendix F). No mention of the instrument height was provided; therefore, we assumed measurements were made at the 10-m level, which is the typical meteorological measurement height. Measurements were reported on 15-minute intervals and included wind speed, wind direction, and atmospheric stability from 2:00 p.m. on May 11, 1969, to 6:00 a.m. on May 12. Mixing layer depths were not provided. Mixing layer depths are calculated hourly within RATCHET at each active meteorological recording station using a methodology described by Zilitinkevich (1972). The RATCHET code also requires default mixing layer depths for each month, stability class, and hour of day. These data were compiled from processed meteorological data taken at the RFP 61-m tower from 1989 to 1993. The calculated or default value is selected based on the relative magnitude of the calculated and default values, stability, season, and time of day. The larger of the calculated default values is selected for the meteorological recording station for the given hour. A multiple linear regression technique is then used to provide a smooth spatial variation in mixing layer depth across the model domain.

Hourly stability classes were calculated separately for the RFP, Jefferson County Airport, and Denver Stapleton International Airport meteorological recording stations using the general classification scheme discussed in Pasquill (1961), Gifford (1961), and Turner (1964). This typing scheme employs seven stability categories ranging from A (extremely unstable) to G (extremely stable) and requires estimates of cloud cover and ceiling height. Cloud cover and ceiling height data for RFP were obtained from Jefferson County Airport while it was operated. During the time the Jefferson County Airport was closed (11:00 p.m. to 5:00 a.m.), data from Denver Stapleton International Airport were used for all stations.

Hourly average wind speed and direction were calculated from the RFP meteorological data using the protocol described in guidance written by the U.S. Environmental Protection Agency (EPA) (EPA 1987). An arithmetic average of the wind direction was computed first, and it was then segregated into 1 of 36, 10-degree increments as required by RATCHET. The average wind speed for the hour was computed by taking the average of the four, 15-minute data segments. No precipitation was recorded in the model domain for the duration of release and subsequent transport and dispersion of material out of the model domain. Meteorological data used in the simulation are summarized in Appendix A.

Atmospheric Transport Model Parameters

This section describes the input parameters we selected for the RATCHET model simulations involving transport and dispersion of plutonium released from the glove box fire in 1969. These parameters include surface roughness length, topography, dry and wet deposition, diffusion coefficients, release parameters (location and height of release), and model control parameters (number of puffs per hour and computational options).

Surface Roughness Length

Roughness elements (such as trees and buildings) and small-scale topographic features (such as rolling hills) have a frictional effect on the wind speed nearest the surface. The height and spacing of these elements determine the frictional effects on the wind. These effects are directly related to transport and diffusion and affect atmospheric stability, wind profiles, diffusion coefficients, and the mixing layer depth. The surface roughness length parameter is used to describe these roughness elements and is a characteristic length associated with surface roughness elements (Table 4). In RATCHET, estimates of the surface roughness length are defined for each node on the environmental grid (Figure 5). In our simulations, we selected a value of 0.6 m to represent residential and urban environs. Farmland, which is predominant in the northeast part of the model domain, was assigned a value of 0.05 m. Range and open land consisting of rolling grass hills were assigned a value of 0.07 m. Nodes that encompass the range and farmland designation were selected based on the topographic contours and land use maps. The foothills and downtown Denver were assigned a value of 2.0 m, and the large open water body (Standley Lake) was assigned a value of 0.001 m.

Table 4. Typical Surface Roughness Lengths for Different Land Use, Vegetation, and Topographic Characteristics^a

Land use, vegetation, and topographic characteristics	Surface roughness length, z_o (m)
Level grass plain	0.007-0.02
Farmland	0.02-0.1
Uncut grass, airport runways	0.02
Many trees/hedges, a few buildings	0.1–0.5
Average, North America	0.15
Average, U.S. Plains	0.5
Dense forest	0.3-0.6
Small towns/cities	0.6–2.5
Very hilly/mountainous regions	1.5+
^a Source: <u>Stull</u> (1988), Figure 9.6.	

Topography

The RATCHET model does not explicitly address terrain differences within the model domain. Instead, topography and topographic effects on transport and diffusion are reflected in the surface roughness lengths and observed wind velocity data that are affected by topographical features. Topography in the model domain (Figure 5) can be characterized by three major features: the north-south trending Colorado Front Range foothills in the western part of the model domain, the southwest to northeast trending Platte River Valley located in the southeast part of the model domain, and rolling hills and flat farmland that are predominant in the central and northeastern part of the model domain. The topography generally slopes east from Rocky Flats, dropping 200 m in elevation to the Platte River Valley. The surface roughness lengths reflect these features as stated in the previous section. Observed meteorological data are lacking in most of the model domain and are woefully inadequate to characterize wind fields in the foothills region. However, meteorological observations at Denver Stapleton International Airport do capture the air movement within the Platte River Valley, which is noticeably different than that at the RFP (DOE 1980). Therefore, to a limited extent, topography is accounted for the model simulation. The use of a complex terrain model would also suffer from the lack of meteorological data, especially in the foothills region. This region is of lesser importance because few receptors were present in the foothills when the RFP was operating.

Dry Deposition and Gravitational Settling

The rate of deposition of small particles on surfaces in the absence of precipitation is proportional to the concentration of material near the surface. The proportionality constant between the concentration in air and the flux to the ground surface is the dry deposition velocity. The current generation of applied models estimates deposition using an analogy with electrical systems as described by Seinfeld (1986). The deposition is assumed to be controlled by a network of resistances, and the deposition velocity is the inverse of the total resistance. Resistances are associated with atmospheric conditions; physical characteristics of the material; and the physical, chemical, and biological properties of the surface. The total resistance in RATCHET is made up of three components: aerodynamic resistance, surface-layer resistance, and transfer resistance. Thus, the dry deposition velocity (v_d , m s⁻¹) is calculated using

$$v_d = (r_s + r_a + r_t)^{-1}$$
 (1)

where

 r_s = surface layer resistance (s m⁻¹) r_a = aerodynamic resistance (s m⁻¹) r_t = transfer resistance (s m⁻¹).

Surface layer resistance and aerodynamic resistance are given by

$$r_a = U(z)/u_*^2 \tag{2}$$

$$r_{\rm S} = 2.6/(0.4 \ u_*) \tag{3}$$

respectively where

U(z) = wind speed (m s⁻¹) measured at height z (m) above the ground

 u_* = frictional velocity (m s⁻¹).

The frictional velocity is given by

$$u_* = \frac{U(z) k}{\ln(z / z_o) - \psi(z / L)}$$
(4)

where

k = the von Karman constant (0.4) z_o = surface roughness length ψ = stability correction factor L = Monin-Obukhov length (m).

The transfer resistance is associated with the characteristics of the depositing material and surface type. In RATCHET, the transfer resistance is used as a mathematical means to place a lower limit on the total resistance. As the wind speed increases, r_s and r_a become small, resulting in unreasonably high deposition velocities. For small particles (<1.0 μ m), a transfer resistance of 100 s m⁻¹ is suggested in RATCHET, and it results in calculated deposition velocities that are consistent with measured data. Harper et al. (1995) estimates deposition velocities for 1 μ m particles and 5 m s⁻¹ wind speed to range from 1.0 × 10⁻² (5th percentile) to 4.1 cm s⁻¹ (95th percentile). The RATCHET-calculated values, assuming a roughness length of 0.05 m and a transfer resistance of 100 s m⁻¹, ranged from 0.66 to 0.75 cm s⁻¹, which is in the range of measured values.

Gravitational settling (v_t) was not originally included in RATCHET and was not included in these simulations. For small particles (~1.0 μ m), gravitational settling is negligible compared to r_s and r_a . Stokes law gives the gravitational settling velocity for particles less than 20 μ m as

$$v_t = \frac{C_c d_p^2 g\rho}{18\mu_{air}} \tag{5}$$

where

 C_c = the Cunningham slip correction factor (dimensionless)

 d_p = physical particle diameter (cm)

g = gravitational acceleration constant (980 cm s⁻²) ρ = particle density (11.46 g cm⁻³ for plutonium) μ_{air} = dynamic viscosity of air (1.78 × 10⁻⁴ g s⁻¹ cm⁻²).

For physical particle diameters less than several microns, the Cunningham Slip correction factor is approximately 1.0. The physical diameter of the particle is related to its aerodynamic equivalent diameter (AED) by

$$d_p = \frac{AED}{\sqrt{\rho_s/\rho_u}} \tag{6}$$

where

 d_p = physical diameter of the particle (μ m) ρ_s = the particle density (11.46 g cm⁻³)

 $\rho_u = \text{unit density of a particle (1 g cm}^{-3}).$

Figure 6 presents gravitational settling velocity as a function of particle size. Plutonium effluent passed through HEPA filtration that was only partially compromised as a result of the fire. This resulted in releases of particles less than 1 μ m in diameter. The median particle size for routine effluent has been estimated to be 0.3 μ m (Voillequé 1999c). Whicker and Schultz (1982) reports that gravitational settling velocities for particles less than 1 μ m are insignificant compared to the other components of deposition. Deposition velocities calculated using Equation (1) ranged from 0.3 to 1.0 cm s⁻¹, for wind speeds ranging from 2.5 to 20 m s⁻¹, roughness lengths from 0.001 to 2 m, and a transfer resistance of 100 s m⁻¹. Note that the gravitational settling velocity for 0.3 μ m particles (\approx 0.006 cm s⁻¹) is insignificant compared to the deposition velocity calculated with Equation (1). For our simulations, we ignored gravitational settling and used a transfer resistance of 100 s⁻¹ m.

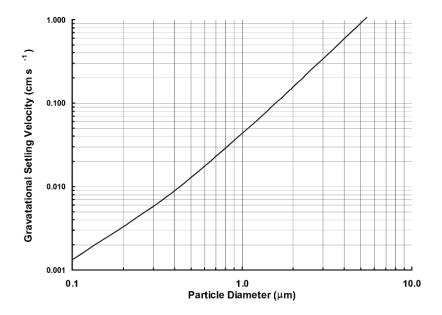


Figure 6. Gravitational settling velocity as a function of physical particle diameter for plutonium (particle density = 11.46 g cm^{-3}).

Wet deposition of small particles in RATCHET is modeled using a washout coefficient and assuming irreversible collection of particles as the precipitation falls through the puffs. Precipitation was not recorded in the model domain during the 1969 fire release event; therefore, this process is irrelevant.

Diffusion Coefficients

In RATCHET, the diffusion coefficients are estimated directly from statistics for atmospheric turbulence. In most cases, the statistics describing atmospheric turbulence (i.e.,

standard deviation of the horizontal and vertical wind direction fluctuations) are not routinely measured at most meteorological recording stations. However, RATCHET makes use of atmospheric conditions that are either measured or calculated from routine meteorological data to estimate the turbulence statistics. The parameters wind speed, atmospheric stability, and surface roughness are used to estimate the turbulence statistics. The general form of the equation used in RATCHET for estimating the horizontal diffusion coefficient (σ_r) for the first hour following release is

$$\sigma_r = 0.5\sigma_v t \tag{7}$$

where

 $\sigma_{v} = \text{crosswind component of turbulence (m s}^{-1})$

t =travel time.

After the first hour, the horizontal diffusion coefficient is given by $\sigma_r = c_{sy} t$, where c_{sy} is a proportionality constant with dimensions of meters per second. Gifford (1983) has shown the value of c_{sy} distributed between 0.14 to 1.4, with a median value of 0.5. For our simulations, we used the median value of 0.5.

The general form of the equation for estimating the vertical diffusion coefficient (σ_z) near the source is

$$\sigma_{z} = \sigma_{w} t f_{z}(t) \tag{8}$$

where

 $\sigma_w = \text{standard deviation of the vertical component of the wind (m s}^{-1})$

 $f_z(t)$ = nondimensional function related to the travel time and turbulence time scale.

As a practical matter, diffusion coefficients in RATCHET are calculated in increments to avoid problems associated with spatial and temporal changes in conditions.

The RATCHET documentation states that the diffusion coefficients implemented in the code are not appropriate for instantaneous puffs. Puff diffusion is defined as when the sampling time is short compared to the travel time of the airborne material (Hanna et al. 1982). Travel times to the eastern margin of the model domain were ~3 hours. If we were interested in the instantaneous concentration at points in the model domain, then puff diffusion coefficients would be required for the simulation. However, we are not calculating instantaneous concentrations, rather time-integrated concentrations (TICs) over the assessment period (16 hours). Because our sampling time (15 hours) is greater than the travel time, the plume diffusion coefficients discussed in this section are appropriate for the simulation.

Source Characterization

Release estimates of plutonium particles were provided by <u>Voillequé</u> (1999a) and are summarized in a previous section or this report. Release estimates were segregated into 15-minute time intervals starting at 2:00 p.m. on May 11, 1969, and continuing until 8:00 p.m. of the same day. For each 15-minute time interval, a nonparametric distribution of release quantities was provided. The distribution was described in terms of percentiles in 5% increments. Originally, RATCHET allowed hourly source updates. The code was modified to allow source updates for every new puff introduced into the model domain. The number of puffs per hour was

set to four so source updates would be recorded every 15 minutes. Each source update consisted of the *quantity* of plutonium released during the source update time increment (15 minutes). Monte Carlo simulations were performed by selecting two random numbers at the beginning of each trial. These random numbers were used to select a percentile from the source term distributions. The same percentile was used for each time interval within the trial, resulting in source release rates that varied only by the total amount of plutonium released during the event (Figure 7).

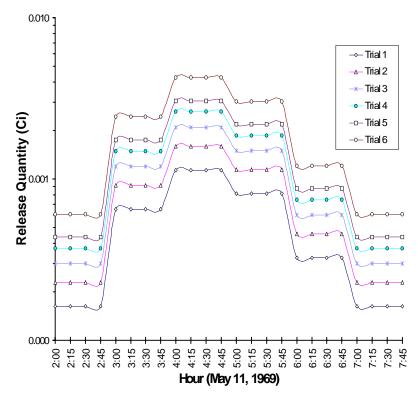


Figure 7. Release from the Building 776/777 roof vents as a function of time for six Monte Carlo trials. For each trial, the timing of the release is not changed; only the total quantity of plutonium released is changed.

Release quantities were provided until 8:00 a.m. May 11, 1969. After that, the source was assumed to go to zero and transport calculations were continued until 5:00 a.m. the following day. Therefore, the RATCHET simulations were performed for 15-hours.

Plutonium suspended as a result of the glove box fire was vented through HEPA-filtered exhaust roof vents (<u>Table 5</u>). Some of the roof vents were inverted "J" vents that directed flow down toward the roof surface and others were rectangular louvered-penthouse vents that vented on all four sides. After the fire, it is believed that many of the inverted "J" roof vents were replaced by louvered-penthouse vents. ChemRisk reports (<u>ChemRisk</u> 1992, Task 3 and 4 Report, Table 5-1) nine roof vents for Building 776/777: six louvered-penthouse vents, two inverted "J," and one stack with a conical stack. This is believed to be the ventilation configuration after the 69 fire. It was reported in a Dow Chemical Company letter (<u>Dow</u> 1970) that four separate ventilation systems were in use in the building. All systems were filtered with HEPA filters

before discharge to the environment. Three booster systems discharged from the processing area, and the forth system filtered the main room area. Effluent from the fire was released through the three booster systems; however Booster System #2 was reportedly clogged with trapped residues from burning plastic and other combustibles. Flow rates for Booster Systems 1 and 3 were 4.93×10^5 and 6.25×10^5 3 d⁻¹, respectively.

Because the effluent flow was directed down toward the building roof, no momentum plume rise was accounted for in the simulation. The vents were spread out over the Building 776/777 roof and releases were assumed to be about the same from each vent. Therefore, an area source was assumed for the geometry of the release. The area source was simulated by modifying the initial diffusion coefficients using a procedure described by Petersen and Lavdas (1986). The initial horizontal diffusion coefficient (σ_r) is the horizontal dimension of the source divided by 4.3, and the initial vertical diffusion coefficient (σ_v) is the height of the source divided by 2.15. For these simulations, we used the 61-m length as the horizontal source dimension and 11.6 m for the building height.

Under routine operations, temperature of the effluent was assumed to be ambient. However, the fire would have heated the effluent, resulting in the potential for buoyant plume rise. However, eyewitness reports (<u>Dow</u> 1970) indicated the following:

The smoke itself was seen at various times to have drifted off at a low angle to the south but for most of the time was described as rolling off the edge of the roof down to the ground on the south side of the building.

Two kinds of smoke were seen. For a brief but undefined period the smoke was black and heavy. This is typical of the fumes from burning Plexiglas and rubber gloves and was seen to be discharged from #1 Booster plenum exhaust. It no doubt was the smoke which carried contamination and lasted for only a brief period. Before, during, and after this period, the major volume of the smoke has been described as light gray-brown. We have generated smoke of this description by testing a section of roof construction material identical to that of Building 776. The gray-brown smoke thus probably came from decomposing the Styrofoam layer in the roof and we have indeed found some areas of decomposed Styrofoam in the Building 776 roof.

Based on these observations, we assumed no buoyant plume rise, and we set the modeled release height to match the height above ground of the Building 776/777 roof. An effluent temperature slightly above the ambient temperature of about 20°C was assumed for the simulation.

Table 5. Release Parameters for Building 776/777 Roof Vents

Parameter	Value
Stack height	11.6 m
Initial $\sigma_{ m r}$	14.1 m
Initial $\sigma_{ m v}$	5.4 m
Effluent temperature	25.0°C
UTM east	482,825 m
UTM north	4,415,740 m

Other Parameters

Several other parameters in RATCHET influence the accuracy of output and computer runtime. These parameters include the number of puffs per hour, minimum time step, puff consolidation, maximum puff radius, and minimum puff concentration at center. We chose the suggested RATCHET default values for all these parameters except minimum time step and minimum concentration at puff centers (Table 6). Accuracy of the simulation can be improved by using a smaller time step. The RATCHET default was 20 minutes, which we reduced to 10 minutes. The minimum concentration at puff centers was reduced from 1×10^{-13} to 1×10^{-15} to allow for plume tracking throughout the model domain. The puff consolidation parameter value combines puffs from the same source when the ratio of the puff centers to the average σ_r is less than the user-input value. The puff consolidation ratio and maximum puff radius (in units of σ_r) were set at RATCHET default values of 1.5 and 3.72, respectively.

Model parameter	Value
Number of puffs per hour	4
Minimum time step	1 minute
Puff consolidation	1.5
Maximum puff radius (in units of σ_r)	3.72
Minimum concentration at puff centers	1×10^{-15}

Table 6. RATCHET Model Control Parameters

Prediction Uncertainty

The uncertainty analysis for the 1969 fire dispersion estimates employed the random sampling features of the RATCHET code because (a) meteorological data were available for the specific release event and (b) random sampling of meteorological input parameters allows for mass balance of the source term with the contaminant mass in the model domain. RATCHET uses random sampling from specified distributions to represent the uncertainty in meteorological data. Specifically, random sampling is limited to wind directions and wind speeds, stability class, Monin-Obukhov length, precipitation rates, and station mixing layer depths. This limitation preserves the physically based correlations among other model parameters and variables. Random sampling of precipitation rates was not used because precipitation did not fall during the event. Uncertainty in the source term and particle size was handled external to the RATCHET code.

Wind Direction Uncertainty. Uncertainty in the wind direction is addressed in RATCHET by sampling from a uniform distribution whose width depends on the measured wind speed. During calm conditions, the width of the distribution is from 0 to 360 degrees. The distribution narrows as the wind speed increases, until the width of the distribution equals the imprecision in the recorded values (a minimum value of 10 degrees). The method used to vary the width of the distribution in RATCHET is based on a procedure described in Schere and Coates (1992). Other sources of uncertainty in wind directions are not considered by the random sampling algorithm in RATCHET. These sources of uncertainty include

- Instrument exposures that may cause observed wind direction to differ systematically from the directions that are representative for the region of measurement
- Changes in wind direction with height that may cause elevated plumes to move in a direction that is different from the one predicted from surface observations.

In reference to the last bullet, <u>Elderkin and Gudiksen</u> (1993) studied several of the WVTS nighttime tests in which additional instrumentation was installed and monitored as part of the Atmospheric Studies in Complex Terrain (ASCOT) program. They found dispersion was controlled by multiple scales of motion, which created interacting layers that varied hourly in three dimensions. Tracer plumes were mostly confined to a stable drainage layer that followed regional flow features, intermittently interrupted by evolving mountain-canyon flows. Based on conventional surface observations, interactions between the surface layer and the mountain-canyon flow layer caused unexpected tracer trajectories. In all atmospheric model simulations performed for this project, we assumed the contaminant plumes remained confined to the surface layers. For the modeling of this event, we believe this is a reasonable assumption because the major releases occurred during daylight hours when mixing heights were high (>2000 m) and unstable (high mixing) conditions existed.

Wind Speed Uncertainty. Wind speeds are recorded in some meteorological records as integer values and in a variety of units. For example, the Denver Stapleton International Airport wind speeds are reported to the nearest whole number in units of knots. This imprecision in wind speed measurements is addressed in RATCHET. RATCHET also addresses the additional uncertainty in wind speeds near and below the threshold.

When random sampling of wind speeds is selected, wind speed is drawn from a uniform probability distribution because with a given wind observation, there is no reason to assume that the actual speed is more or less likely to be in any part of the range of values. The width of the distribution is two reporting units. For example, if the measured wind speed is 5 m s⁻¹, then the width of the distribution is from 4 to 6 m s⁻¹. When a calm wind is reported, a wind speed between 0 and 1 m s⁻¹ is used.

Stability Uncertainty. Atmospheric stability is a fundamental concept in meteorology, but it cannot be calculated directly from the available meteorological data. Therefore, stability must be estimated from the limited data that are available.

Methods of estimating stability classes proposed by <u>Gifford</u> (1961), <u>Pasquill</u> (1961), and <u>Turner</u> (1964) are based on data that are available in routine meteorological observations, such as those taken at airports. These methods form the basis of the procedures that the National Climatic Data Center uses to estimate stability classes from climatological data (<u>Hatch</u> 1988).

Golder (1972) compares stability class estimates made at five locations using the method proposed by Pasquill and Turner's variation. The results of this comparison, presented in Golder (1972, Figure 3), show reasonable agreement among the hourly stability class estimates. However, other studies, such as the study in Luna and Church (1972), show that these stability classes have a much wider range of uncertainty when attempting to estimate turbulence characteristics related to diffusion.

RATCHET allows the user to specify the uncertainty associated with stability class estimates. This uncertainty is represented by a set of seven conditional cumulative frequency distributions—one conditional cumulative frequency distribution for each stability class. The cumulative frequency distribution represents the possible actual stability class for the one

reported stability class. To do this, we employed two different methods of calculating stability: (1) the method described in <u>Turner</u> (1964) and used to define nominal values for stability class and (2) the lateral turbulence and wind speed method (standard deviation of the horizontal wind direction fluctuations) as described in <u>EPA</u> (1987). Stability classes were calculated for 5 years of meteorological data taken at the RFP between 1989 and 1993. This was the same meteorological data set used for routine release and transport calculations (<u>Rood</u> 1999b). Conditional cumulative frequency distributions were input through a file containing seven records, one for each stability class (<u>Table 7</u>). Each record contains seven values that are the cumulative probability that the actual stability class is the same as the reported stability class.

For example, the probability that a reported stability class of 1 is actually 1 is 0.934 (see Table 7, line one column 2). The probability that a reported stability class of 1 may actually be 2 is 0.961 - 0.934 = 0.027. The probability that a reported stability class of 1 may actually be 3 is 0.988 - 0.961 = 0.027. The probability that a reported stability class of 1 may actually be 4 is 1.00 - 0.988 = 0.012 and so on.

Stability	Cumula	tive frequen	cy that the a	ctual stabili	ty class is ≤	the reported	stability class
class	1	2	3	4	5	6	7
1	0.934	0.961	0.988	1.000	1.000	1.000	1.000
2	0.565	0.819	0.927	0.996	0.998	1.000	1.000
3	0.268	0.409	0.704	0.955	0.981	1.000	1.000
4	0.072	0.113	0.213	0.895	0.975	1.000	1.000
5	0.000	0.000	0.000	0.597	0.994	1.000	1.000
6	0.000	0.000	0.000	0.339	0.629	1.000	1.000
7	0.000	0.000	0.000	0.110	0.279	1.000	1.000

Table 7. Conditional Cumulative Frequency Distributions for Stability Class

Monin-Obukhov Length. Stability classes are discrete estimates of atmospheric stability. However, boundary-layer similarity theory uses the reciprocal of the Monin-Obukhov length, which is a continuous variable, to represent stability. Figure 2.4 in the RATCHET documentation (Ramsdell et al. 1994), which is based the Golder (1972) paper, provides a basis for converting stability class to Monin-Obukhov length (Figure 8). When random sampling of the reciprocal Monin-Obukhov is selected, RATCHET obtains an appropriate value as needed from a uniformly distributed range of values. The upper and lower bounds of the range are computed from the surface roughness and stability class.

Mixing Layer Depth. RATCHET computes mixing layer depth from the friction velocity and Monin-Obukhov length. For stable conditions, the mixing layer depth is given by

$$H = k \left(\frac{u_* L}{f}\right)^{1/2} \tag{9}$$

where

H = mixing layer depth (m)

 $k = \text{von Karman constant (} \sim 0.4 \text{ dimensionless)}$

 $u_* =$ friction velocity (m s⁻¹) L =Monin-Obukhov length (m) f = Coriolis parameter (s⁻¹).

<u>Pasquill and Smith</u> (1983) indicates that constant values in the range of 0.2 to 0.7 have been suggested in place of the von Karman constant. <u>Weil</u> (1985) suggests constant values in the range of 0.4 to 0.7. For neutral or unstable conditions, the mixing layer depth is estimated using

$$H = \frac{\beta u_*}{f} \tag{10}$$

where β = a dimensionless constant. Zilitinkevich (1972) assumes β is equal to k, while Pasquill and Smith (1983) suggests a value in the 0.2 to 0.3 range. Other researchers suggest its range is from 0.15 to 0.25 (Panofsky and Dutton 1984). When random sampling of the mixing depth is selected, RATCHET samples from uniform distributions to obtain the value of k and β . The range of k is fixed between 0.2 and 0.7 and the range of β is fixed between 0.15 and 0.3.

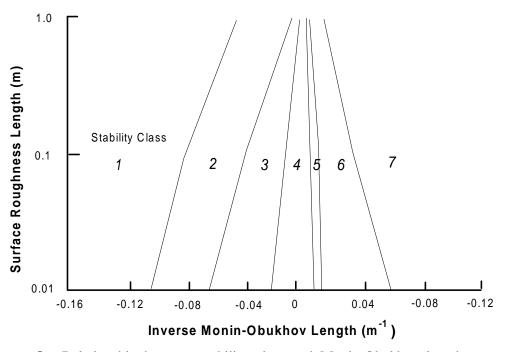


Figure 8. Relationship between stability class and Monin-Obukhov length as a function of surface roughness length (redrawn from <u>Ramsdell et al.</u> 1994).

Integration of Uncertainty Analysis into Model Predictions. The uncertainty analysis required a Monte-Carlo simulation that coupled RATCHET atmospheric transport simulations with distributions of the 1969 fire source term. Calculations were performed using a FORTRAN pre-and post-processor program that (1) sampled the release rate from distributions of these quantities, (2) wrote RATCHET input files, (3) executed the RATCHET simulation, and (4) extracted and stored results. The source release rate and meteorological parameters were considered independent of one another.

Output consisted of 1000 trials of 15-hour average atmospheric concentrations and deposition at the 2295 receptor nodes in the model domain. Average concentrations were converted to *TICs* by multiplying by the number of hours in the simulation (15 hours). The *TIC* values were used in the intake and risk calculations.

Predicted Air Concentrations

Average and time-integrated plutonium concentrations were calculated throughout the model domain using the source term developed by <u>Voillequé</u> (1999a) and the atmospheric modeling procedure described in previous sections. Distributions of 15-hour time-integrated air concentrations for 15 receptor locations (illustrated in Figures 9 and 10) have been described in terms of the 5th, 50th, and 95th percentile values of the distribution of predicted air concentration values (<u>Table 8</u>). We used these statistics to describe the concentration distributions; they were not used in the risk calculation. Instead, we used the actual distributions comprising 1000 RATCHET realizations to calculate plutonium intake and risk to the receptors. The 15 receptor locations chosen for risk calculations represented individuals from each of the major population centers in addition to receptors placed at locations of high concentration in the model domain. The maximum 15-hour average air concentration at the 95%, 50%, and 5% level for the 15 receptor locations was 0.67, 0.17 and 0.03 pCi m⁻³, respectively, and it occurred at the west entrance to the RFP. The maximum location west of the RFP reflects the predominance of easterly winds during the highest releases from the fire.

Fifteen-hour average concentrations in the *entire* model domain (including onsite locations) ranged from a minimum of 0 to a maximum of $0.11 \, \mathrm{pCi} \, \mathrm{m}^{-3}$ at the 5% level and 5.2×10^{-4} to 14 pCi m⁻³ at the 95% level. Figure 9 shows the number of simulations where the 15-hour average concentration was greater than 0 for 1000 Monte Carlo trials. The 95% contour line (950 of the simulations had a 15-hour average concentration greater than zero) encompassed an area that includes most of the model domain excluding only the upper northwest corner (northwest of the City of Boulder). Unlike the routine release evaluation (Rood 1999b) where nonzero concentrations were calculated for all receptor nodes, the fire has an additional component of uncertainty; that is, the probability that the plume even reached the receptor.

The median estimate (50th percentile value) of the 15-hour average concentration is illustrated in Figure 10. The dispersion pattern depicted represents the 50th percentile concentration estimate at each of the 2295 receptor nodes. Dispersion patterns are typical of what we would expect for releases from the RFP during daylight hours and under upslope conditions (Crow 1974). Upslope conditions are a result of daytime heating and typically result in easterly winds that prevail during the daylight hours. The transition from upslope to downslope conditions occurs during the evening, and transition from downslope to upslope occurs during the morning. During evening hours under stable conditions, cool air near the surface drains from the Denver metropolitan area down the Platte River Valley (which flows to the northeast) and out to the plains. During daylight hours and after surface heating has eliminated the cooler surface layer, the downslope conditions cease. This is followed by a brief period of relatively calm winds, which in turn is followed by return of air up the valley or upslope conditions. Typically, unstable conditions exist that result in rapid mixing to elevations >2000 m above the ground surface. Analysis of hourly dispersion patterns indicated the plume initially moved west of the RFP till about 10:00 p.m. This was followed by a change to downslope conditions and a reversal of the plume trajectory to the east. The highest releases occurred from about 3:00 to 6:00 p.m.; consequently, the highest concentrations were predicted to occur west of the RFP.

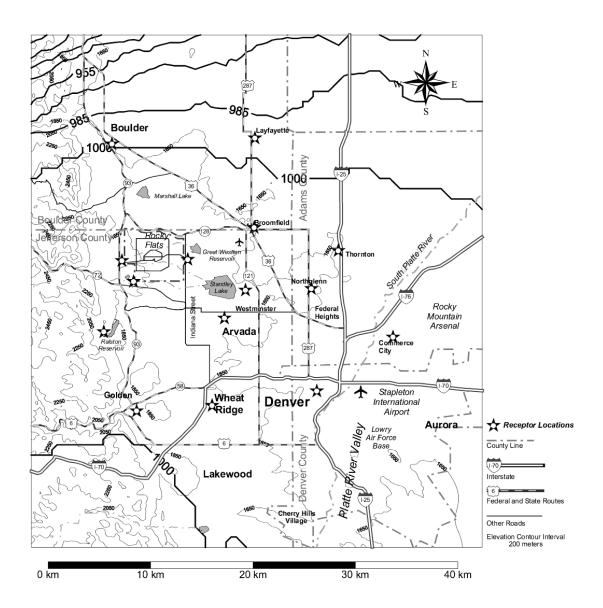


Figure 9. Number of Monte Carlo trials where the 15-hour average plutonium concentration in air was greater than 0 for 1000 trials. For example, the 950 contour line indicates the location where 95% of the simulations had 15-hour average concentrations greater than 0. Receptor locations used in the risk calculations are indicated by a star.

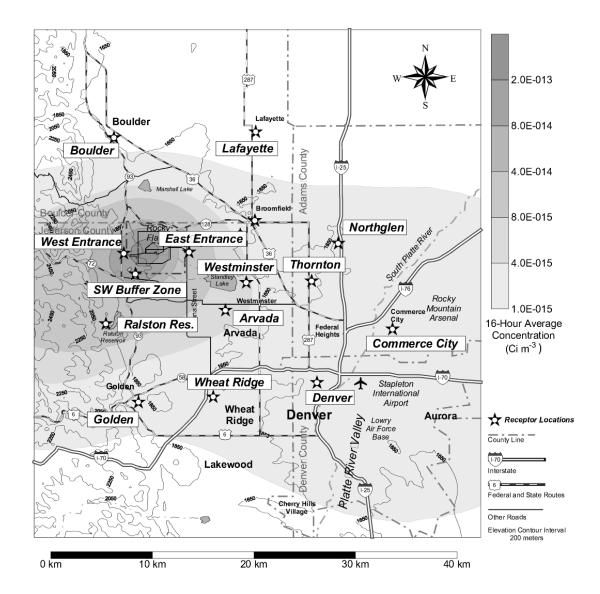


Figure 10. Predicted 15-hour average plutonium concentration in air at the 50 percentile level. Concentrations were based on 1000 Monte Carlo trials. Receptor locations that are used in the risk calculations are indicated by a star and identified by the text box.

Table 8. Predicted 15-hourAverage Concentrations for Plutonium in Air at 15 Receptor Locations in the Model Domain

			15-hou	15-hour average concentration				
Receptor location		_		(pCi m ⁻³)				
UTM E (m)	UTM N (m)	Description	5%	50%	95%			
495821	4412980	SW Buffer Zone	2.0×10^{-2}	1.3×10^{-1}	5.2×10^{-1}			
486115	4415130	East Entrance	2.6×10^{-3}	2.0×10^{-2}	1.1×10^{-1}			
479684	4415010	West Entrance	3.1×10^{-2}	1.7×10^{-1}	6.7×10^{-1}			
477917	4408070	Ralston Reservoir	2.4×10^{-3}	1.5×10^{-2}	5.7×10^{-2}			
489696	4409400	Arvada	9.5×10^{-4}	4.6×10^{-3}	1.5×10^{-2}			
488491	4400870	Wheat Ridge	1.8×10^{-4}	2.0×10^{-3}	6.9×10^{-3}			
491747	4412140	Westminster	9.5×10^{-4}	4.2×10^{-3}	1.3×10^{-2}			
492593	4418250	Broomfield	2.9×10^{-4}	2.2×10^{-3}	8.5×10^{-3}			
500804	4415990	Thornton	2.4×10^{-4}	1.8×10^{-3}	5.8×10^{-3}			
498184	4412290	Northglenn	5.6×10^{-4}	2.6×10^{-3}	7.6×10^{-3}			
478709	4426410	Boulder	1.9×10^{-5}	7.4×10^{-4}	5.3×10^{-3}			
492674	4426980	Lafayette	1.2×10^{-5}	4.6×10^{-4}	3.0×10^{-3}			
481126	4400350	Golden	1.2×10^{-4}	1.8×10^{-3}	7.8×10^{-3}			
506152	4407550	Commerce City	5.3×10^{-4}	2.5×10^{-3}	6.6×10^{-3}			
498712	4402290	Denver	4.5×10^{-4}	2.6×10^{-3}	7.2×10^{-3}			

In Phase I of this study, the 1969 fire plumes had both a westerly and strong easterly component. We did not observe this phenomenon in our simulations because the majority of the Phase II release was postulated to occur over a relatively short period of time (3 hours) during which the winds were predominately from the northeast. In Phase I, the release was assumed to occur over a period of time that encompassed the change in wind direction observed at 10:00 p.m.

EXPOSURE SCENARIOS AND RISK CALCULATIONS

One of the key parts of the Rocky Flats dose reconstruction work is calculating health impacts to people living in the surrounding area from materials released during RFP past operations. Dose reconstruction uses a pathways approach to study the potential radiation doses and health risks of these past releases on the surrounding communities. The pathways approach begins with learning what kinds of and how much materials were released from a facility (source terms) and ends with estimating the health impacts these releases had on the residents in the area. Mathematical models described in the previous sections were used to model the transport of plutonium released from the site to the surrounding communities. In this section, we present the method for calculating health impacts (incremental lifetime cancer incidence risk) to people living offsite from exposure to these releases.

The risk to a person from exposure to the plutonium released depends upon a number of factors, such as

• Where the person lived and worked in relation to the RFP

- Did the person live near the RFP during the 1969 fire
- The age and gender of the person
- Lifestyle (that is, did the person spend a great deal of time outdoors or doing heavy work on a farm).

Although it is not realistic to calculate individual risks for every resident who may have lived or worked in the Rocky Flats area during its operational history, it is not credible to calculate a single risk that applies to all residents. To consider the many factors that influence exposure, we developed profiles, or exposure scenarios, of hypothetical but realistic residents of the RFP area for which representative risk estimates could be made. Each scenario represents one individual. These scenarios incorporate typical lifestyles, ages, genders, and lengths of time in the area. The scenarios also specify home and work locations. These scenarios can help individuals determine risk ranges for themselves by finding a lifestyle profile that most closely matches their background. The scenarios are not designed to include all conceivable lifestyles of residents who lived in this region during the time of RFP operations. Rather, they provide a range of potential profiles of people in the area.

We calculated the risks to hypothetical individuals from plutonium released to the air during the 1969 fire for seven hypothetical exposure scenarios at 15 separate locations in the model domain (Table 9). Locations were selected to include all major population centers and to intercept the plume path where the maximum concentration occurred. As discussed earlier, direct inhalation was the only exposure pathway considered in this assessment. Ingestion of plutonium in water, food, and soil are potential pathways that could have been considered in more detail. However, plutonium compounds are very insoluble and tend to adhere to soil, making them relatively immobile and not readily taken up by plants or accumulated in the edible portions of animal products. Phase I results (ChemRisk 1994d) indicated direct inhalation to be the dominant pathway of exposure during the early period of RFP operations (1952–1970). For the later years (1970-1989), soil ingestion and inhalation of resuspended contaminated soil become a significant component of the total dose because of the accumulation and build up of deposited plutonium in soil and smaller airborne releases. This report deals only with risks from the 1969 fire, which are dominated by direct inhalation of airborne activity released by the fire. Risk from inhalation of resuspended soil activity was addressed in a separate report (Rood and Grogan 1999) that considers all sources of offsite plutonium contamination.

Table 9. Exposure Scenario Descriptions

Exposure		Year of	
scenario	Gender	birth	Locations
Rancher	Male	1925	Southwest corner of cattle fence
Office worker	Female	1941	Denver
Housewife	Female	1928	Arvada, Wheat Ridge, Westminster, and Broomfield
Laborer	Male	1943	West Entrance, Ralston Reservoir Thornton, Boulder, and Lafayette
Infant	Female	1957	Northglenn
Child	Female	1953	Commerce City
Student	Female	1949	Golden

Exposure scenarios for the seven hypothetical receptors described in Table 9 were organized according to occupational and nonoccupational activities. Occupational activities included work, school, and extracurricular activities away from the home. Nonoccupational activities included time spent at home doing chores, sleeping, and leisure activities (such as watching television). In these calculations, the receptor was assumed to perform occupational and nonoccupational activities at the same location. The age of the receptor during which exposure occurred was also considered when calculating risk. All scenarios assumed the individual was exposed for the duration of the 1969 fire event.

Breathing Rates and Time Budgets

Each exposure scenario was divided into three types of activities: sleeping, nonoccupational, and occupational activities. For the infant and child scenario, occupational and nonoccupational activities are irrelevant; instead, activities were divided into sleeping and two other activities based on the child's age. For the infant, the other two activities were awake sedentary and awake active. For the child scenario, the two other activities were time spent at home (indoors and outdoors) and at preschool or day care.

For each activity, time spent at four different exercise levels was assigned. These exercise levels were resting, sitting (sedentary), light exercise, and heavy exercise. Some examples of light exercise are laboratory work, woodworking, housecleaning, and painting. Heavy exercise corresponds to occupations such as mining, construction, farming, and ranching. For each exercise level, an age- and gender-specific breathing rate was assigned. Breathing rates (Table 10) for persons age 8 and higher were obtained from Roy and Courtay (1991) and for children age 0–7 from Layton (1993).

Time budgets for various receptor activities were also based on Roy and Courtay (1991) (Table 11), but they were modified to fit specific exposure scenarios and the timing of the fire event. The fraction of time spent at a specific exercise level while engaged in a given activity was assigned based on the nature of the activity. For example, the fraction of time spent at the resting exercise level while the receptor slept would be 1.0 and the other exercise levels would be 0. A weighted-average breathing rate was then applied to each activity based on the number of hours spent at each exercise level. For some scenarios (housewife, retiree, and laborer), nonoccupational activities were separated into those performed indoors and those performed outdoors. Although no distinction was made between indoor and outdoor air concentrations, exercise levels for indoor and outdoor activities differed. A time-weighted average breathing rate that included indoor and outdoor activities was calculated and applied to nonoccupational time.

Time-weighted average breathing rates were calculated for the three activities for which each receptor was assumed to be engaged. The time-weighted average breathing rate is given by

$$WBR_{j} = \sum_{i=1}^{4} BR_{i} f_{i,j}$$
 (11)

where

 WBR_j = time-weighted average breathing rate for the j^{th} activity (m³ h⁻¹)

 BR_i = breathing rate for the i^{th} exercise level (m³ h⁻¹)

 $f_{i,j}$ = fraction of time spent at the i^{th} exercise level for the j^{th} activity.

Table 10. Breathing Rates for Various Exercise Levels as Reported in Roy and Courtay (1991) and Layton (1993)

		Exercise level					
		Resting	Sitting	Light	Heavy		
Gender	Age	$(m^3 h^{-1})$	$(m^3 h^{-1})$	$(m^3 h^{-1})$	$(m^3 h^{-1})$		
Male	30-60	0.45	0.54	1.50	3.00		
Female	30-60	0.32	0.39	1.26	2.70		
Male	18	0.50	0.60	1.58	3.06		
Female	18	0.35	0.42	1.32	1.44		
Male	16	0.43	0.52	1.52	3.02		
Female	16	0.35	0.42	1.30	2.70		
Male	15	0.42	0.48	1.38	2.92		
Female	15	0.35	0.40	1.30	2.57		
Male	14	0.41	0.49	1.40	2.71		
Female	14	0.33	0.40	1.20	2.52		
Male	12	0.38	0.47	1.23	2.42		
Female	12	0.33	0.39	1.13	2.17		
Male	10	0.31	0.38	1.12	2.22		
Female	10	0.31	0.38	1.12	1.84		
Male	8	0.29	0.39	1.02	1.68		
Female	8	0.29	0.39	1.02	1.68		
Male	3–7	0.24	0.29	0.72	1.68		
Female	3–7	0.23	0.27	0.68	1.59		
Male	0–3	0.19	0.23	0.58	1.35		
Female	0–3	0.14	0.17	0.45	1.02		
Average, male ^a	8–17	0.37	0.45	1.28	1.49		
Average, female ^a	8–17	0.33	0.40	1.18	2.25		

^a The average female breathing rate from age 8–17 was used for the student.

Table 11. Time Budgets and Weighted Breathing Rates for the Exposure Scenarios

		Fraction of time spent at a given activity level					Weighted breathing rate
Scenario	Activity	Resting	Sitting	Light	Heavy	Hours	$(m^3 h^{-1})$
Rancher	Occupational	0.00	0.00	0.25	0.75	5.0	2.625
	Nonoccupational	0.00	0.50	0.38	0.13	5.0	1.208
	Sleeping	1.00	0.00	0.00	0.00	5.0	0.450
	Weighted daily average						1.428
Office worker	Occupational	0.00	0.25	0.75	0.00	5.0	1.042
	Nonocuupational	0.00	0.50	0.38	0.13	5.0	1.004
	Sleeping	1.00	0.00	0.00	0.00	5.0	0.324
	Weighted daily average						0.790
Housewife	Occupational	0.00	0.13	0.75	0.13	6.0	1.331
	Indoor nonoccupational	0.00	0.50	0.38	0.13	2.0	1.004
	Outdoor nonoccupational	0.00	0.38	0.50	0.13	2.0	1.113
	Total nonoccupational	0.00	0.44	0.44	0.13	4.0	1.058
	Sleeping	1.00	0.00	0.00	0.00	5.0	0.324
	Weighted daily average						0.923
Laborer	Occupational	0.00	0.125	0.50	0.375	5.0	1.943
	Indoor nonoccupational	0.00	0.5	0.375	0.125	2.5	1.208
	Outdoor nonoccupational	0.00	0.5	0.25	0.25	2.5	1.208
	Total nonoccupational	0.00	0.50	0.31	0.19	5.0	1.395
	Sleeping	1.00	0.00	0.00	0.00	5.0	1.301
	Weighted daily average						1.231
Infant	Awake—sedintary	0.00	0.71	0.14	0.14	5.0	0.334
	Awake—active	0.00	0.00	1.00	0.00	5.0	0.447
	Sleeping	1.00	0.00	0.00	0.00	5.0	0.144
	Weighted daily average						0.308
Child (2–6)	Indoor (home)	0.00	0.50	0.42	0.08	3.0	0.549
	Outdoor (home)	0.00	0.00	0.67	0.33	4.0	1.040
	Total home					7.0	0.794
	Indoor (school)	0.00	0.80	0.20	0.00	1.0	0.351
	Sleeping	1.00	0.00	0.00	0.00	7.0	0.228
	Weighted daily average						0.501
Student (7–18)	Indoor home	0.00	0.44	0.56	0.00	3.0	0.829
	Outdoor home	0.00	0.00	0.25	0.75	4.0	1.979
	Total home	0.00	0.22	0.40	0.38	7.0	1.404
	Indoor school	0.00	0.75	0.25	0.00	1.0	0.591
	Outdoor school	0.00	0.00	0.25	0.75	1.0	1.979
	Total school	0.00	0.38	0.25	0.38	2.0	0.000
	Sleeping	1.00	0.00	0.00	0.00	6.0	0.326
	Weighted daily average						0.824

To summarize, three activities were defined for each exposure scenario: sleeping, occupational, and nonoccupational activities. The location of exposure for occupational and nonoccupational activities was assumed to be the same for all receptors. Four different exercise levels, each with an assigned breathing rate, were distinguished: resting, sitting, light exercise, and heavy exercise. The breathing rate during a given activity was the time-weighted average breathing rate of the four exercise levels.

At the time of the major releases from the fire (2:00–5:00 p.m.), most people would be engaged in occupational activities. Breathing rates while doing nonoccupational activities, such as watching television or sleeping, are substantially less than while awake. Ideally, the calculation of plutonium intake would be described by

$$I = \int_0^t C(t) BR(t) dt \tag{12}$$

where C(t) = the plutonium concentration as a function of time and BR(t) = the breathing rate as a function of time. It was not practical to provide the function C(t) for each Monte Carlo trial; instead, the TIC value was provided $[\int C(t) dt]$. Therefore, the breathing rate applied to the intake calculation had to account for not only the activities performed by the receptor but the fact that most of the intake occurred over the first several hours of the release event. Consequently, for the adult receptors, we assumed only 5 hours were spent at the sleeping breathing rate. We also assumed the remainder of the hours was spent doing occupational and nonoccupational activities because it is likely these receptors were awake during the major releases from the event. For the infant scenario, we assumed an equal amount of time for all three activity levels. For the child scenario, we assumed 7 hours of the exposure time was spent sleeping and the remainder was spent awake. For the student, who represents a person 7–18 years old, we assumed 6 of the 15 exposure hours were spent sleeping.

Plutonium Intake Calculation

Calculating the incremental lifetime cancer incidence risk involved three steps:

- 1. Calculate the *TIC* in air at the point of exposure
- 2. Calculate the amount of plutonium inhaled by the receptor
- 3. Multiply the plutonium intake by a risk coefficient that relates the incremental lifetime cancer incidence risk to the amount of plutonium inhaled.

The 15-hour average concentrations reported earlier were calculated from *TIC* values by dividing the *TIC* by the simulation time (15 hours). Uncertainty in risk estimates includes uncertainty in the *TIC* and risk coefficients. Receptor behavior patterns (i.e., the time spent doing different activities at different exertion levels) and their physical attributes (body weight and breathing rate) were considered fixed quantities. The amount of plutonium inhaled by a receptor for the 15-hour exposure period is given by

$$I = TIC \frac{\left(WBR_1 T_1 + WBR_2 T_2 + WBR_3 T_3\right)}{ED}$$

$$\tag{13}$$

where

Ι

= intake of plutonium by the receptor for the exposure period (Ci)

TIC = time-integrated concentration (Ci-h m⁻³)

 $WBR_{1,2,3}$ = time-weighted average breathing rate for occupational, nonoccupational, and

sleeping activity (m³ h⁻¹)

 $T_{1,2,3}$ = hours during the exposure period for occupational, nonoccupational, and sleeping

activity (h)

ED = exposure duration (15 hours).

The subscripts 1, 2, and 3 refer to occupational, nonoccupational, and sleeping activity, respectively. Note that the values for *WBR* in <u>Table 11</u> are weighted toward the time spent doing occupational and nonoccupational activity to account for exposure to the bulk of the release during waking hours.

Risk Coefficients

Calculating the lifetime cancer incidence risk requires estimates of risk coefficients. Risk coefficients relate the lifetime risk of cancer incidence to the amount of plutonium inhaled. Plutonium risk coefficients were developed in Phase II of the study and are documented in Grogan et al. (1999).

The principal plutonium isotopes of concern at Rocky Flats are ²³⁹Pu and ²⁴⁰Pu, which have long half-lives of 24,065 years and 6537 years, respectively. Plutonium emits alpha particles that are relatively heavy and slow, thus, creating short, dense ionization trails. Alpha particles have such weak penetration abilities that they can be blocked by a piece of paper or the dead, outer layers of the skin. As a result, the major danger from plutonium comes from having it inside your body. For residents in the vicinity of Rocky Flats, plutonium is most likely to have entered the body from breathing air that contained plutonium particles released from the site. After inhalation, plutonium enters the blood and about 80% is transported to the bone or liver where it is retained for years. Following inhalation, the four most highly exposed tissues are bone surface, lung, liver, and bone marrow. These tissues account for more than 97% of the total dose received by infants and adults alike. The dose per unit activity inhaled varies for these four tissues (see Table 12). Furthermore, the dose per unit activity (dose conversion factor) also varies depending on the particle size distribution of the inhaled plutonium aerosol (Table 12). The particle size distribution of the inhaled plutonium aerosol was assumed to have an activity median aerodynamic diameter (AMAD) of 1 µm and a GSD of 2.5 (Table 12). This particle size distribution accounts for routine vent and stack effluents that were effectively filtered which result in aerosols with an AMAD of ~0.3 μm, and the larger particles that would have been released when filter leakage occurred. The inhaled plutonium is assumed to be in the oxide form.

The incidence of health effects depends on the amount of dose received. Two main classes of health effects are induced by ionizing radiation: deterministic and stochastic effects. Deterministic effects most often follow acute, high dose exposure. The severity of the effect increases with dose above the threshold dose. Below the threshold dose, the effect is not evident; however, subtle minor effects may occur. Deterministic effects cause direct damage to tissues and include effects that most often occur within days to weeks after exposure. For example, these effects can cause reddening of the skin, cataracts, hair loss, sterility, and bone marrow depression after external irradiation. After inhalation of plutonium, deterministic effects may include radiation pneumonitis, pulmonary fibrosis, and lymphopenia, but these conditions occur only

after very high doses. The threshold dose for most deterministic effects is at least 0.5 Gy delivered in a short time, and many are much higher (NCRP 1991). For the releases of plutonium that occurred from the site, doses to individuals in the Rocky Flats area were well below the threshold doses. Therefore, deterministic health effects were not possible.

Table 12. Plutonium Inhalation Dose Conversion Factors for a 1-μm AMAD Aerosol with a GSD of 2.5^a

Cancer site	Dose conversion factor (μGy Bq ⁻¹) ^b			
Lung	4.4 (1.9)			
Liver	2.0 (3.0)			
Bone	9.0 (3.0)			
Bone marrow	0.46 (3.0)			
^a ICRP (1995)				
^b Geometric mean (geometric standard deviation).				

Stochastic effects are assumed to occur randomly at all dose levels, including the lowest doses. The frequency of stochastic effects is dependent on the dose, and the effects usually occur at long intervals after exposure. In a large population exposed to low doses, only a few of the exposed individuals will be affected, most will not. The two principal types of stochastic effects are induced cancer and genetic effects. For exposure to plutonium, the risk of induced cancer is the health effect of most concern; in particular, lung cancer, liver cancer, bone cancer, and leukemia (bone marrow exposure) because these are the tissues that receive the highest doses. Genetic effects are not an important risk for plutonium exposures because (a) people exposed to radiation are several times more likely to be affected by an induced cancer than to transmit genetic effects to their children and (b) the plutonium doses to the gonads (ovaries or testes) are small compared to other organs of the body (40 times less than the lung). Therefore, we did not consider them further.

The alpha particles emitted from plutonium are densely ionizing, and the linear energy transfer (LET) to the tissue is high over the short range (about 40 μ m) of the alpha particles (thus, the name high-LET radiation). Other radiations, such as gamma rays and x-rays, are less densely ionizing and are termed low-LET radiations. The biological effects of low-LET radiation are better known than those of high-LET radiation. The differences between radiation types are important to the analysis because high-LET radiations are more biologically effective (cause more damage) per unit of dose than low-LET radiations. This difference in effectiveness is usually described by the relative biological effectiveness (RBE), which is the ratio of doses from two different radiations to produce the same type and level of biological effect.

Inhalation of plutonium results in the exposure of organs to high-LET radiation. While a few human populations have been exposed directly to large amounts of plutonium and some populations to other radionuclides that emit alpha particles, more groups have been exposed to low-LET gamma radiation and have been evaluated in more epidemiologic detail. In addition, studies of cancer in animals exposed to both types of radiation and laboratory studies of cellular and other biological endpoints can be used to support human studies. These different sources of information were used in this phase of the study to develop four independent approaches to estimate the risk of cancer because of radiation doses from plutonium deposited in the organs of the human body (Grogan et al. 1999). Three approaches used epidemiologic studies of human populations to derive dose-response relationships, and the fourth approach used dose-response

relationships from controlled animal experiments. The four independent approaches were used to derive, where possible, risk coefficients for each organ of interest. The coefficients from the different approaches were then combined by weighting each according its intrinsic merit to produce a single risk coefficient with uncertainties for each organ of interest.

The overall mortality risk estimate for each cancer site was adjusted by the lethality fraction to provide lifetime risk estimates for cancer incidence. The influence of gender and age was accounted for in the analyses (see <u>Grogan et al.</u> [1999] for details). The data allowed a distinction to be made between the risks and uncertainties to those under 20 years of age at exposure and those 20 and older. The data did not warrant a more detailed analysis. For this reason, the risk coefficients for persons under 20 years of age were applied to the infants and children in the seven hypothetical exposure scenarios.

The GM (50th percentile) and GSDs of the cancer incidence risk coefficient distributions are listed in Table 13. The units reported in Grogan et al. (1999) have been changed from risk per 100,000 persons per unit of activity in kilobecquerels (kBq) to risk per 10,000 persons per unit of activity in microcuries (μ Ci). These numbers indicate the median number of cases of cancer (fatal and nonfatal) that would be expected to result from 10,000 people all inhaling 1 μ Ci of $^{239/240}$ Pu particles with the defined particle size distribution. The particle size distribution of the effluent was assumed to be in the range of HEPA-filtered effluent (<1.0 μ m). Therefore, the risk coefficients for 1- μ m particles were used in the risk calculations

Table 13. Lifetime Cancer Incidence Risk Per 10,000 Persons Per 1 μCi of Inhaled ^{239/240}Pu for 1 μm AMAD Particles^a

1-μm AMAD particles (GSD = 2.5)						
Cancer site	Gender	Age under 20	Age 20 and older			
Lung	Male	206 (3.5)	210 (3.4)			
-	Female	206 (3.5)	210 (3.4)			
Liver	Male	92 (5.2)	49 (5.2)			
	Female	45 (5.4)	23 (5.4)			
Bone surface	Male	16 (9.5)	8.0 (9.3)			
	Female	8.0 (10)	4.0 (10)			
Bone marrow	Male	2.4 (6.1)	2.3 (6.3)			
	Female	2.4 (6.1)	2.3 (6.3)			
^a Geometric mean (geome	etric standard devia	tion).				

Risk Calculations

Plutonium intake (Equation [13]) was multiplied by the risk coefficients (Table 13) to yield lifetime cancer incidence risk (R_i) for each organ of interest.

$$R_j = I \times RC_j \tag{14}$$

where

 R_i = lifetime cancer incidence risk for the j^{th} organ

I = plutonium intake (Ci)

 $RC_{,j}$ = risk coefficient for the j^{th} organ (Ci⁻¹).

Risk coefficients for the different organs were assumed to be correlated. For each Monte Carlo trial, a standard normal deviate was generated and stored. These deviates were then used to determine a risk coefficient for each organ of interest using Equation (15), substituting the appropriate GM and GSD of the specific organ.

$$RC = \exp(d\ln(GSD) + \ln(GM)) \tag{15}$$

The total lifetime cancer incidence risk from all organs was calculated by summing the risk across all four organs during each Monte Carlo trial.

Risk calculations were performed using a FORTRAN programs that (a) read *TIC* values from the RATCHET output file, (b) computed plutonium intake, (c) sampled risk coefficients and calculated risk, and (d) stored and processed output. FORTRAN routines for generating random numbers and normal deviates were adapted from <u>Press et al.</u> (1992). The output distributions provided in this report were generated from 1000 trials.

LIFETIME CANCER INCIDENCE RISK ESTIMATES

Incremental lifetime cancer incidence risks were expressed in terms of percentiles of the cumulative density function (Tables $\underline{14}$ – $\underline{16}$ and $\underline{Appendix}$ B). At all percentile levels, the receptor with the maximum total (all organs) risk in the model domain was the laborer located at the west entrance, followed by the rancher located at southwest buffer zone. Total risk for the laborer at the west entrance ranged from 1.2×10^{-8} at the 2.5% level to 4.9×10^{-7} at the 97.5% level. Using this scenario as an example, the uncertainty in these risk estimates may be interpreted as follows:

- There is a 95% probability that the incremental lifetime cancer incidence risk was between 1.2×10^{-8} (2.5% value) and 4.9×10^{-7} (97.5% value).
- There is a 2.5% probability that the incremental lifetime cancer incidence risk was greater than 4.9×10^{-7} (97.5% value) and a 2.5% probability the risk was lower than 1.2×10^{-8} (2.5% value)

We may also interpret this to mean, given an exposure history and lifestyle similar to that of the laborer, there is a 97.5% probability that the model-predicted number of cancer cases attributed to inhalation of plutonium originating from the 1969 fire release would be no greater than 5 persons in a population of 10 million similarly exposed individuals. In all cases, the organ with the greatest risk was the lung, followed by the liver, bone surface, and bone marrow.

An almost infinite number of possible exposure scenarios can be defined; in most cases, the risks associated with each scenario will differ. However, the maximum risks will probably be bounded by the risks associated with the laborer scenario located at the west entrance to the RFP. These scenarios may be considered to represent the maximum exposed individuals in the model domain because they were placed at the point of highest concentration outside the RFP cattle fence. In addition, the laborer was assumed to be working during the time of major releases, thereby, maximizing his breathing rate during the releases from the fire.

The median (50%) risk estimates calculated for Phase II are lower than those calculated by Phase I. Geometric mean risk for maximum exposed individual in Phase I (located 3 mi from the RFP) was 3×10^{-7} while the 50% risk value for the maximum exposed individual in Phase II was 8.6×10^{-8} (about a factor of 3.5 difference). However, median Phase I airborne activity

concentrations were lower than those calculated in Phase II. Based on the isopleth maps in the Phase I Task 6 Report (ChemRisk 1994c), the maximum 17.5-hour average concentration for an offsite location was around 0.05 pCi m⁻³ (0.88 pCi-h m⁻³ *TIC*). For Phase II, the maximum 15-hour average concentration at the 50% level was 0.17 pCi m⁻³ (2.6 pCi-h m⁻³ *TIC*) which is about a factor of 3 higher than Phase I. Because the airborne concentrations in Phase II were higher than Phase I, the compensating differences existed in calculating dose and risk between the two phases.

Phase I determined preliminary risk estimates and, therefore, used a less rigorous approach than Phase II to estimate radiation dose and cancer incidence risk. In Phase I, the plutonium inhalation dose conversion factor was represented by a uniform distribution between the values given in ICRP Report 56 (ICRP 1990) for slightly soluble (Class W) and insoluble (Class Y) forms of plutonium. The dose conversion factor ranged from 1.2×10^{-4} Sv Bq⁻¹ to 8.4×10^{-5} Sv Bq⁻¹. In Phase II, uncertainty was explicitly accounted for in the inhalation dose conversion factor. This resulted in significantly different values for the conversion from intake to risk compared to Phase I. Additionally, the plutonium released during the fire was postulated in Phase II to be plutonium oxide and highly insoluble. Dose conversion factors are lower for insoluble plutonium than the soluble form. Lifetime cancer incidence risk coefficients with uncertainties for exposure to plutonium were estimated for the principal organs of concern: lung, liver, bone and bone marrow. In contrast, a single value for the whole body risk estimate of 7.3 percent per sievert was assumed in Phase I. For comparison, the Phase II estimated lifetime cancer incidence risks are within the U.S. Environmental Protection Agency point of departure for acceptable lifetime cancer incidence risk of 1 in 1,000,000 to 1 in 10,000 people.

Table 14. Lifetime Incremental Cancer Incidence Risk at the 2.5% Level for 1969 Fire Releases

				Bone	Bone	
Receptor	Location	Lung	Liver	surface	marrow	Total
Rancher	SW Cattle Fence	5.9×10^{-9}	1.4×10^{-9}	2.9×10^{-10}	6.6×10^{-11}	7.6×10^{-9}
Rancher	East Entrance	8.5×10^{-10}	2.0×10^{-10}	4.2×10^{-11}	9.5×10^{-12}	1.1×10^{-9}
Laborer	West Entrance	9.0×10^{-9}	2.1×10^{-9}	4.5×10^{-10}	1.0×10^{-10}	1.2×10^{-8}
Laborer	Ralston Reservoir	7.3×10^{-10}	1.7×10^{-10}	3.6×10^{-11}	8.2×10^{-12}	9.5×10^{-10}
Housewife	Arvada	2.0×10^{-10}	2.2×10^{-11}	4.9×10^{-12}	2.3×10^{-12}	2.3×10^{-10}
Housewife	Wheat Ridge	3.2×10^{-11}	3.6×10^{-12}	7.7×10^{-13}	3.6×10^{-13}	3.7×10^{-11}
Housewife	Westminster	2.0×10^{-10}	2.2×10^{-11}	4.7×10^{-12}	2.2×10^{-12}	2.3×10^{-10}
Housewife	Broomfield	4.7×10^{-11}	5.3×10^{-12}	1.1×10^{-12}	5.3×10^{-13}	5.4×10^{-11}
Laborer	Thornton	5.7×10^{-11}	1.3×10^{-11}	2.8×10^{-12}	6.4×10^{-13}	7.4×10^{-11}
Infant	Northglenn	3.9×10^{-11}	8.4×10^{-12}	1.9×10^{-12}	4.5×10^{-13}	4.9×10^{-11}
Laborer	Boulder	2.4×10^{-12}	5.7×10^{-13}	1.2×10^{-13}	2.7×10^{-14}	3.1×10^{-12}
Laborer	Lafayette	2.2×10^{-12}	5.3×10^{-13}	1.1×10^{-13}	2.5×10^{-14}	2.9×10^{-12}
Student	Golden	2.4×10^{-11}	2.9×10^{-12}	6.5×10^{-13}	1.6×10^{-13}	1.7×10^{-11}
Child	Commerce City	5.4×10^{-11}	1.2×10^{-11}	2.6×10^{-12}	6.3×10^{-13}	6.8×10^{-11}
Office Worke	r Denver	7.5×10^{-11}	8.4×10^{-12}	1.8×10^{-12}	8.4×10^{-13}	8.6×10^{-11}

Table 15. Lifetime Incremental Cancer Incidence Risk at the 50% Level for
1969 Fire Releases

				Bone	Bone	
Receptor	Location	Lung	Liver	surface	marrow	Total
Rancher	SW Cattle Fence	5.8×10^{-8}	1.4×10^{-8}	2.9×10^{-9}	6.5×10^{-10}	7.5×10^{-8}
Rancher	East Entrance	9.0×10^{-9}	2.1×10^{-9}	4.4×10^{-10}	1.0×10^{-10}	1.2×10^{-8}
Laborer	West Entrance	6.6×10^{-8}	1.5×10^{-8}	3.3×10^{-9}	7.4×10^{-10}	8.6×10^{-8}
Laborer	Ralston Reservoir	5.8×10^{-9}	1.3×10^{-9}	2.9×10^{-10}	6.4×10^{-11}	7.5×10^{-9}
Housewife	Arvada	1.3×10^{-9}	1.5×10^{-10}	3.2×10^{-11}	1.5×10^{-11}	1.5×10^{-9}
Housewife	Wheat Ridge	5.8×10^{-10}	6.4×10^{-11}	1.4×10^{-11}	6.5×10^{-12}	6.6×10^{-10}
Housewife	Westminster	1.2×10^{-9}	1.4×10^{-10}	3.0×10^{-11}	1.4×10^{-11}	1.4×10^{-9}
Housewife	Broomfield	6.4×10^{-10}	7.1×10^{-11}	1.5×10^{-11}	7.1×10^{-12}	7.3×10^{-10}
Laborer	Thornton	6.9×10^{-10}	1.6×10^{-10}	3.4×10^{-11}	7.8×10^{-12}	9.0×10^{-10}
Infant	Northglenn	2.5×10^{-10}	5.4×10^{-11}	1.2×10^{-11}	2.9×10^{-12}	3.2×10^{-10}
Laborer	Boulder	2.9×10^{-10}	6.7×10^{-11}	1.4×10^{-11}	3.2×10^{-12}	3.7×10^{-10}
Laborer	Lafayette	1.8×10^{-10}	4.1×10^{-11}	8.8×10^{-12}	2.0×10^{-12}	2.3×10^{-10}
Student	Golden	4.5×10^{-10}	9.7×10^{-11}	2.2×10^{-11}	5.2×10^{-12}	5.7×10^{-10}
Child	Commerce City	3.8×10^{-10}	8.3×10^{-11}	1.8×10^{-11}	4.4×10^{-12}	4.9×10^{-10}
Office Worker	r Denver	6.4×10^{-10}	7.1×10^{-11}	1.5×10^{-11}	7.1×10^{-12}	7.3×10^{-10}

Table 16. Lifetime Incremental Cancer Incidence Risk at the 97.5% Level for 1969 Fire Releases

				Bone	Bone	
Receptor	Location	Lung	Liver	surface	marrow	Total
Rancher	SW Cattle Fence	3.1×10^{-7}	7.2×10^{-8}	1.5×10^{-8}	3.4×10^{-9}	4.0×10^{-7}
Rancher	East Entrance	6.4×10^{-8}	1.5×10^{-8}	3.2×10^{-9}	7.1×10^{-10}	8.3×10^{-8}
Laborer	West Entrance	3.8×10^{-7}	8.8×10^{-8}	1.9×10^{-8}	4.2×10^{-9}	4.9×10^{-7}
Laborer	Ralston Reservoi	1.0×10^{-8}	7.0×10^{-9}	1.5×10^{-9}	3.3×10^{-10}	3.9×10^{-8}
Housewife	Arvada	5.2×10^{-9}	5.8×10^{-10}	1.2×10^{-10}	5.8×10^{-11}	5.9×10^{-9}
Housewife	Wheat Ridge	2.5×10^{-9}	2.7×10^{-10}	5.9×10^{-11}	2.8×10^{-11}	2.8×10^{-9}
Housewife	Westminster	4.6×10^{-9}	5.1×10^{-10}	1.1×10^{-10}	5.1×10^{-11}	5.2×10^{-9}
Housewife	Broomfield	3.1×10^{-9}	3.4×10^{-10}	7.3×10^{-11}	3.4×10^{-11}	3.5×10^{-9}
Laborer	Thornton	2.6×10^{-9}	6.1×10^{-10}	1.3×10^{-10}	2.9×10^{-11}	3.4×10^{-9}
Infant	Northglenn	8.2×10^{-10}	1.8×10^{-10}	3.9×10^{-11}	9.6×10^{-12}	1.0×10^{-9}
Laborer	Boulder	2.7×10^{-9}	6.3×10^{-10}	1.3×10^{-10}	3.0×10^{-11}	3.5×10^{-9}
Laborer	Lafayette	1.8×10^{-10}	4.1×10^{-11}	8.8×10^{-12}	2.0×10^{-12}	2.3×10^{-10}
Student	Golden	1.5×10^{-9}	3.5×10^{-10}	7.4×10^{-11}	1.7×10^{-11}	1.9×10^{-9}
Child	Commerce City	2.4×10^{-9}	5.3×10^{-10}	1.2×10^{-10}	2.8×10^{-11}	3.1×10^{-9}
Office Worke	r Denver	1.2×10^{-9}	2.6×10^{-10}	5.7×10^{-11}	1.4×10^{-11}	1.5×10^{-9}

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